Numerical simulation and experimental study of the acetylene hydrogenation reaction

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Abstract. A mathematical model of the acetylene hydrogenation reaction is constructed. Numerical modeling and experimental studies of the hydrogenation of acetylene in a reactor with a catalyst based on open cell foam material have been carried out. The results of numerical modeling and experimental studies are in good agreement. The calculation results show that with increasing temperature in the reactor, the mass content of acetylene decreases.

1. Introduction

Open cell foam materials are the subject of much research because of their unique properties. They are characterized by a large contact surface area and high porosity; this creates a low pressure drop and provides high energy efficiency compared to traditional systems with a compacted layer [1-3]. Currently, metallic porous materials are widely used as multifunctional heat exchangers [4] and compact radiators for microelectronic devices [5-6], due to their heat transfer properties [7-8]. Also, heat-conducting porous materials have been proposed as an effective solution for the intensification of non-adiabatic catalytic processes in tubular reactors.

In chemical engineering, porous materials are widely used as catalyst supports. In addition to chemical properties, during chemical reactions, the technological parameters and the geometric structure of fixed-bed catalytic reactors, including porous materials, play a decisive role. For stable and reliable operation of reactors in industrial conditions, it is necessary to correctly set the operating parameters for monitoring the thermo-fluid dynamics in the reactor. The mode of operation of catalytic reactions related to a chemical reaction or their field of application determines the characteristic requirements and limitations of reactors, including size, throughput, heat transfer uniformity, pressure drop, and selectivity. Numerical simulations can characterize fixed-bed catalytic reactors for industrial applications, or optimize and improve them. For exothermic reactions, solid porous structures exhibit interesting results. In [9], ceramic porous materials, honeycombs, and bulk spheres were compared. The authors found that the main advantage of porous materials is their radial mixing characteristics. Radial heat transfer from the pipe wall to the reactor and vice versa is a key aspect in chemical engineering. In [10], porous metal granules with solid porous structures were compared in terms of pressure drop and heat transfer characteristics. However, there is little research in the literature on the advantages and disadvantages of porous materials used in fixed-bed reactors.

Ethylene is an essential component of the petrochemical industry. An important task in the production of ethylene is its purification from side components, for example, acetylene. In this case,
purification can be carried out by hydrogenation, when acetylene is converted to ethylene. However, not only acetylene but also the ethylene, which hydrogenates to ethane, can be hydrogenated. Thus, the process should take place in the presence of a catalyst with selectivity parameters allowing acetylene hydrogenation, but not promoting ethylene hydrogenation.

2. Problem formulation and methods

2.1. Reactor model

Let us consider the problem of hydrogenation of acetylene in a reactor with an open cell foam catalyst. Palladium acts as a catalyst. In laboratory conditions, experiments were conducted on a cylindrical reactor with a diameter of 2 cm and a height of 70 cm. In the middle of the reactor is a foam material coated with palladium 5.5 cm high. The parameters of the experimental sample of foam material: cell diameter – 2-3 mm, window diameter – 1.5-2 mm.

To carry out the calculations, we will construct a model of a reactor with an open cell foam catalyst (figure 1).

![Figure 1. The open cell foam catalyst model.](image)

A gas fraction is passed through the reactor, containing 75% of the volume of ethylene, 0.7% of the volume of acetylene, 0.98% of the volume of hydrogen, 23.32% of the volume of argon. The volumetric gas flow through the reactor is 5000 ml/h.

We consider the hydrogenation reaction of acetylene

$$C_2H_2 + H_2 \rightarrow C_2H_4 + 176.8 \text{kJ/mol.}$$  

(1)

The experiments were carried out in the gas temperature range of 30-600°C. Acetylene conversion ranged from 23% to 100%. To calculate the conversion, we will use the following ratio

$$C = \frac{1 - C_{\text{out}}}{C_{\text{in}}} \times 100\%,$$  

(2)
2.2. Reactive gas flow mathematical model
Let write the equations of conservation of mass, energy, and momentum for a stationary flow of a multicomponent gas phase. Equations are solved in the ANSYS Fluent CFD package.

The mass conservation equation
\[ \nabla \cdot (\rho \vec{v}) = 0, \]  
where \( \rho \) is the density, \( \vec{v} \) is the velocity.

The momentum conservation equation
\[ \nabla \cdot (\rho \vec{v}) = -\nabla p + \nabla \cdot \vec{T} + \rho \vec{g}, \]  
where \( p \) is the pressure, \( \vec{T} \) is the stress tensor.

The mass conservation equation for the \( i \)-th component of the gas mixture
\[ \nabla \cdot (\rho_i \vec{v}) = -\nabla \cdot \vec{J}_i + R_i, \]  
where \( Y_i \) is the mass fraction for the \( i \)-th component of a gas mixture, \( J_i \) is the diffusion flux of \( i \)-th species, which arises due to gradients of concentration and temperature.

The energy conservation equation
\[ \nabla \cdot (\rho h) + \nabla \cdot (\vec{J}_q) = \frac{\partial p}{\partial t} + \vec{T} : \vec{v}, \]  
where \( h = \sum h_i \) is the gas enthalpy, and
\[ \vec{J}_q = \lambda \nabla T + \sum_{i=1}^{N} h_i \vec{J}_i, \]  
where \( \lambda \) is the thermal conductivity of mixture, \( N \) is a number of components in the mixture.

For chemical reactions, the term \( R_i \) in equation (6) is
\[ R_i = M \sum_{r=1}^{N_R} R_{i,r}, \]  
where \( M_i \) is the molecular mass for \( i \)-th component, \( N_R \) is a number of reactions involving the \( i \)-th component of the mixture.

\[ R_{i,r} = \nu_j \eta_j \prod_{j=1}^{N} C_j^{\eta_j}, \]  
where \( \nu_j \) is the stoichiometric coefficient, \( C_j \) is the concentration of the \( j \)-th component of mixture, \( \eta_j \) is the rate exponent for reactant \( j \)-th species in reaction, \( k_r \) is the reaction rate constant.
\[ k_r = A \exp^{-E_r/RT}, \].
where $A_r$ is the pre-exponential coefficient, $E_r$ is the activation energy.

2.3. Numerical simulation method

For the numerical solution of the problem, the entire area under consideration is divided into finite elements of a triangular shape, the dimensions of which are sufficient to determine the specific factors of the phenomenon under study. We specify conditions at the boundaries of the considered region.

The considered flow of the gas (at a temperature of 30-60°C) is considered to be a multicomponent ideal incompressible gas consisting of raw materials (ethylbenzene and water vapor) and reaction products (styrene and hydrogen). The physicochemical properties of the components are given according to the polynomial dependence on temperature.

To numerically solve the problem of hydrodynamics and heat and mass transfer in a reactor, we specified conditions at the boundaries of the region under consideration. Boundary conditions were established in accordance with the operating mechanisms of the calculated reactor and the selected solver used at all boundaries of the computational domain. The impermeable wall condition has been established on all impermeable surfaces. At the gas flow supply section, the conditions for the gas mass flow rate were set in the mixer model; for steam is 5000 ml/h at 30-60°C. Physico-chemical properties of gases are automatically recalculated depending on the temperature in polynomial relationships. On the site in the reactor model for the gas outlet, the conditions of external pressure outside the considered region were set.

3. Results

Numerical calculations were carried out for the operation of the reactor at a temperature of 30, 40, 50, and 60 °C. Figures 2-5 show the acetylene mass fraction fields in the reactor. It is seen that with increasing temperature, the mass content of acetylene decreases.

![Figure 2. C2H2 mass fraction, T=30°C.](image)

![Figure 3. C2H2 mass fraction, T=40°C.](image)
We compare the results of numerical modeling of gas purification from acetylene with experimental results. Figure 6 presents the results of comparing the mass content of acetylene at the outlet of the reactor. The repeating nature of the two curves is visible.

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References
[1] Giani L, Groppi G and Tronconi E 2005 Ind. Engin. Chem. Res., 44(14) 4993
[2] Richardson J T, Peng Y and Remue D 2000 Appl. Catalysis A: General 204 (1) 19
[3] Lu T J, Stone H A and Ashby M F 1998 Acta materialia 46 (10) 3619
[4] Haack D P, Butcher K R., Kim T and Lu T J 2001 ASME Congress Proceedings 1
[5]  Banhart J. 2001 Prog. Mater. Sci. 46 (6) 559
[6]  Solovev S A, Khusainov R R and Nasretdinova D R 2019 IOP Conf. Ser.: Materials Sci. Eng. 618 012096
[7]  Ghosh I 2009 Int. J. Heat and Mass Transfer, 52 (5-6) 1488
[8]  Coquard R, Loretz M and Baillis D 2008 Adv. Engin. Mat. 10 (4) 323
[9]  Patcas F C, Garrido G I and Kraushaar-Czarnetzki B 2007 Chem. Engin. Sci. 62 (15) 3984
[10]  Kolaczkowski S T, Awdry S, Smith T, Thomas D, Torkuhl L and Kolvenbach R 2016 Catalysis Today 273 221