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Numerical simulation of oxidative regeneration of a spherical catalyst layer*

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Abstract: The article presents the results of numerical simulation of a spherical catalyst layer with single-stage kinetics. The model is described by partial differential equations. The diffusion-reaction equations correspond to the material balance of the reaction's gas phase in the grain pores. The inhomogeneous equation of heat conductivity corresponds to the heat balance. The decrease in the mass fraction of coke sediments on the catalyst grain is described by an ordinary differential equation. Heat and mass transfer in the reaction mixture layer is taken into account. The computational algorithm is constructed using the integro-interpolation method. The results of the software are the profiles of the distribution of temperature fields across the catalyst layer, the dynamics of the mass fractions of the reaction mixture in the catalyst layer, the average values of the concentrations of substances in the catalyst grain.

Keywords: mathematical modeling, integro-interpolation method, diffusion-reaction equations, heat conductivity equation, oxidative regeneration.

The issue of resource conservation is one of the most important questions in our days. An urgent task of industrial chemistry is to study the possibility of reuse of the catalyst within the framework of this problem.

During the reactions, the catalyst inevitably loses its effectiveness, including by reducing the active surface due to the accumulation of coke on the surface and in the grains' pores [1]. One of the methods of restoring catalytic activity is the oxidative regeneration – the burning of coke sediments with oxygen-containing gas [2]. The main disadvantage of the reduction process is the prolonged exposure to high temperature on the catalyst, which violates its physico-chemical properties, structure, and also leads to an increase in the fragility of the material [3]. It is obvious that regeneration needs strict temperature control while reducing the burning time. This leads to the need to predict the flow of the process under various technological conditions.

Mathematical modeling is an effective mean of solving such problems [4]. Modern computing capabilities make it possible to carry out serial calculations fairly quickly with various input data of a mathematical model [5, 6].

The mathematical model of oxidative regeneration of the catalyst layer is two-phase – the gas and solid phases are described separately. It is a system of partial differential equations [7, 8]. The system includes equations of material balance by the number of substances involved in the reaction. The material balance of the gas phase in the catalyst

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grain (oxygen and carbon dioxide) is described by diffusion equations with a source term corresponding to chemical transformations – diffusion-reaction equations. The decrease of coke mass fraction on the catalyst grain is described by an ordinary differential equation. The model also includes the equations of the thermal balance for the catalyst grain – the heat exchange of the gas and catalyst layers, the heating of the grain due to chemical reactions and its propagation along the reactor's length is taken into account. The calculations are performed under the assumption of grain isothermicity, which greatly simplifies the model and the computational algorithm for it [9]. In turn, the material and thermal balances of the gas in the catalyst layer are described by the heat and gas transfer equations, respectively, taking into account the heat and mass transfer of the gas and the catalyst layer.

The initial boundary conditions for the system describe the entry of gas into the catalyst layer and heat and mass transfer at the grain boundary. The difference scheme for the model is based on the integro-interpolation method and has an explicit-implicit character [10].

The computational algorithm is implemented in C++. The results of the software are the profiles of the distribution of temperature fields across the catalyst layer, the dynamics of the mass fractions of the reaction mixture in the catalyst layer, the average values of the concentrations of substances in the catalyst grain.

The pictures fig. 1Fig. 1:-2Fig. 2: present changes of characteristics. The initial values is follow. Oxygen in the catalyst pores and in the reaction gas is 100%, catalyst's temperature is 273 K (0°C), temperature of reaction mixture is 793 K (520°C).

Different lines on the graphs correspond to different lengths of the catalyst layer in meters (transcript in the right columns).

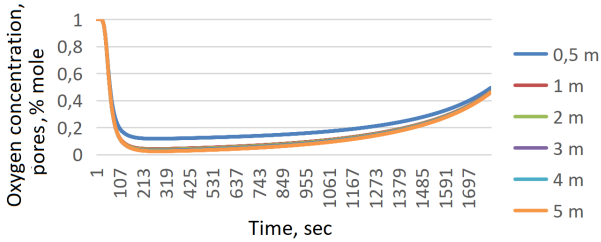

Fig. 1: Oxygen in pores, mole fractions

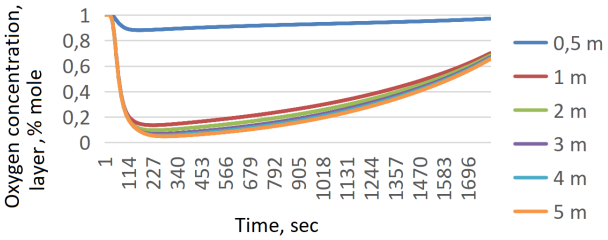

Fig. 2: Oxygen in gas, mole fractions

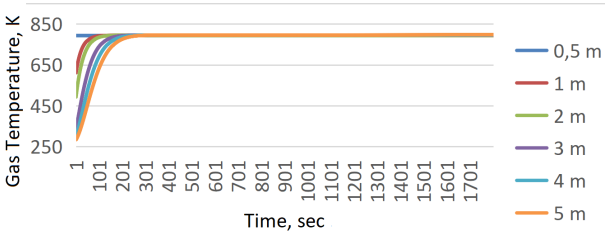

Fig. 1: Gas temperature, K

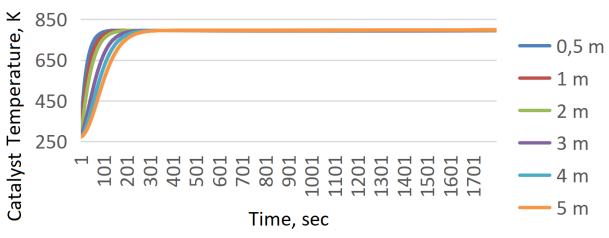

Fig. 2: Catalyst temperature, K

The temperature of the grain is compared with the temperature of the gas over time.

Reactions go fast enough from the first minute. Oxygen in the grain's pores burns out, while the mass transfer between the grain and the gas layer is not fast enough for smooth burning. This leads to low oxygen concentrations both in the pores and in the gas layer at the first minutes.

The oxygen concentration reaches the initial value by the end of the burn (about thirty minutes) due to the fact that the coke has burned out and the reaction is not going on.

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