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Computer simulation of flow reactors*

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Abstract. The numerical 3D modeling of the gas-dynamic reactants flows of a flow reactor and their mixing that was made with the FLUENT software package is discussed. The presence of the modes where a reaction zone with a high content of C_2 hydrocarbons is localized in the center of reactor presented. A good relation between computational modeling and experimental data has been obtained.

1. Introduction

Endothermic chemical processes are carried out in the reactors with energy input to a reaction zone. Such reactors are distinguished by an energy source, which can be represented by an electric arc [1–4], the heat from combustion of a part of feedstock with oxygen in the reactor [5], cyclic heating of a catalyst [6], an inert gas at a high temperature [7], or reactor tubes [8,9]. A wide variety of the heating methods and endothermic chemical processes implies special-purpose designs of chemical reactors: turbulent flow reactors, reactors with shock waves in a tube, and plasma reactors [10–14].

One of the main problems is optimization of reactants residence time in the reaction zone to prevent involvement of target products in secondary reactions. Thus, thermal dehydrogenation of C_2–C_4 hydrocarbons yielding ethylene and propylene is characterized by a high reactivity of these products at the temperatures of pyrolysis. This imposes specific constraints on the reactor design.

The use of laser radiation for heating the reaction medium makes possible the direct input of energy to a gas with the power density of 100 W/cm^2, which exceeds by half, i.e., by two orders of magnitude, the values attained in reactors with metal walls. This circumstance allows decreasing the time of reactants pyrolysis to milliseconds. The possibility of using the laser energy as a powerful energy source for chemical reactors has given impetus to the scientists for studying endothermal chemical processing. There are many different trends initiated in studying gas-core reactions [15–29, 32], preparation of catalysts [20], polymer films synthesis [22], heat-and-mass transfer, interaction between laser radiation and gas medium, the effect of laser radiation energy on chemical reactions equilibrium [24–28] during 70–80s of

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the XXth century. At the power density below 10^5–10^6 W/cm^2, energy absorption and reaction occurrence at atmospheric and higher pressures follow the thermal mechanism [29]. A necessary condition for application of laser radiation is that a reaction medium comprises a component whose absorption spectrum contains bands coinciding with the region of laser generation. In this case, the absorbed energy of laser radiation is transmitted at collisional relaxation to a reaction medium, thus heating it. When studying the kinetics process, it is important to provide the gas-phase occurrence of the reactions. A necessary thermal insulation of a reaction zone can be provided by a buffer gas supercharging at the sites of laser radiation input and by a relatively cold mixture of reactants in the rest near-wall region of convectively cooled reactor. For the cylindrical geometry, such a mode can be implemented via a smaller section of laser beam as compared to the reactor section.

This paper presents results of numerical modeling of a flow reactor with the laser input of energy for endothermic reactions accompanied with ethane pyrolysis.

The pilot prototype of the reactor was tested with the FLUENT software package.

2. Computer simulation

An optimal kinetics scheme was computed using the ChemPAK. The reactions proceeded under homogeneous conditions at atmospheric pressure in the reactor with the reaction mixture components heated by the laser energy flux delivered directly into the gas. Ethylene, being among the prime reaction products, served as a laser energy absorber. This created the mode of 'energetic catalysis'. In this case, increasing energy absorption in the volume is associated with an increased content of the reaction product.

Mixing gas flows in the 3D reactor geometry was calculated with the FLUENT software package. For all flows, the FLUENT solves the conservation equations for mass and momentum. A full description of the mathematical model that was used in the FLUENT is presented in [32]. Many tests with a numerical model with different parameters of reactant and buffer gas flows, radiation model were carried out. Figures 1 and 2 show cross-sections of a chemical reactor. The design of the chemical reactor is fully described in [31].

Results of numerical modeling in the Figure 1 were obtained using a constant absorption coefficient (the laser energy absorbtion by reactant gases). This model does not equal to experiment because the absorption coefficient depends on C_2H_4 concentration. But we can rapidly obtain results of modeling the gas flows (2–3 hours of the FLUENT running on a standard double cores processor) with a 20–30 % error per experiment. We can observe the

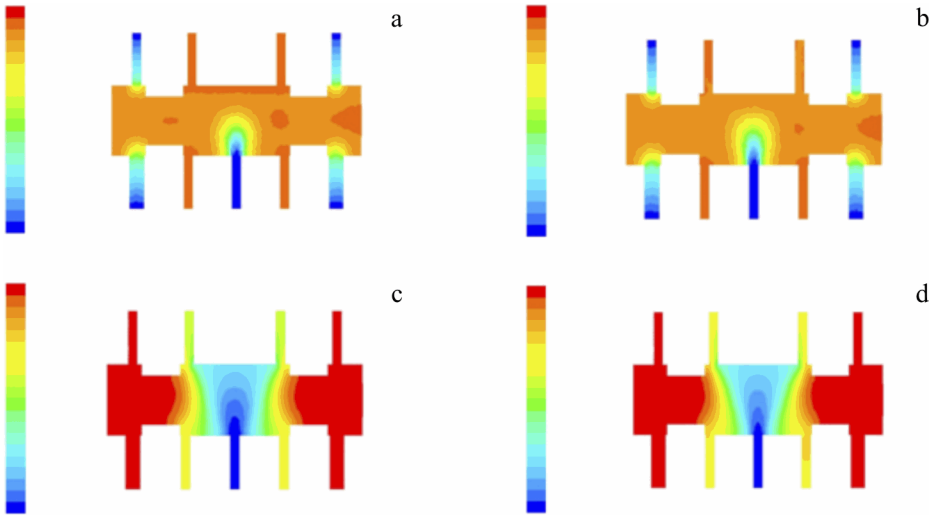

Figure 1. Contours of temperature (a,b) and Ar mass fraction (c,d) for C_2H_6–C_2H_4 mass flow rates 1.1 L/h (a,c) and 2.1 L/h (b,d) and Ar mass flow rates 2.5 L/h (a,c) and 3.45 L/h (b,d) at time 200 s

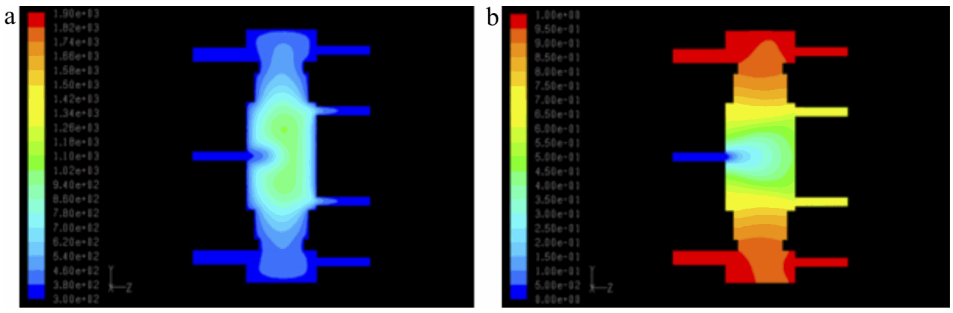

Figure 2. Contours of temperature (a) and Ar mass fraction (b) for C_2H_6–C_2H_4 mass flow rate 1.1 L/h and Ar mass flow rate 2.5 L/h at time 200 s

Argon block reactant gases in the middle of the reactor. By this is meant that the reaction products do not fall out on the end windows, and these windows will be clear during the chemical process.

The results in Figure 2 have been obtained with a variable absorption coefficient that depends on C_2H_4 concentration. The user defines a function for a variable absorption coefficient developed for the FLUENT. This scheme is closer to the experiment. But the time of calculation is twice as large as a constant coefficient. The temperature in the reaction zone is 1100–1200 K that is sufficient for starting chemical reactions. The concentration of reactants and temperature at windows are too low for starting chemical reactions.

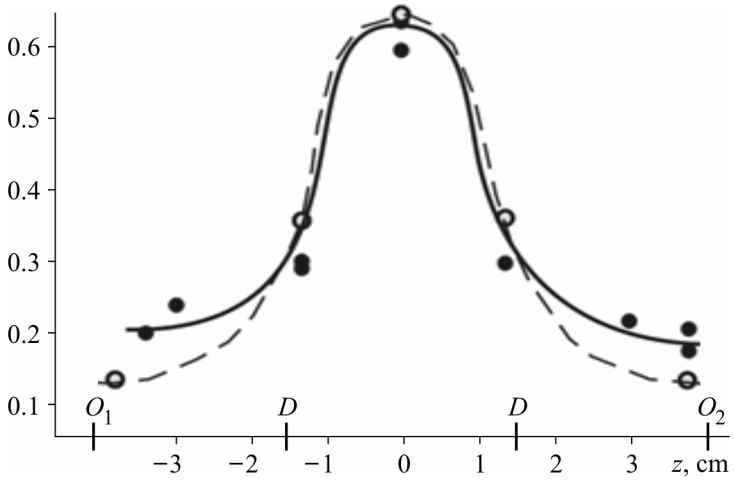

Figure 3. Distribution of hydrocarbons over the length of reactor in the experiments: O_1 and O_2 are reactor faces, D are diaphragm boundaries in the reaction zone. Dark points mark experimental data. Hollow points mark calculated data obtained with the FLUENT software [31]

Figure 3 shows a good relation between experimental data and numerical modeling using the ChemPAK and the FLUENT software packages.

3. Conclusion

A flow reactor with laser energy input providing homogeneous conditions for a chemical process occurrence has been devised for studying gas-phase reactions. The 3D modeling of gas flow mixing was carried out using the FLUENT program package for the reactor with geometrical dimensions corresponding to the pilot sample. The qualitative agreement between the calculated and the experimental data was obtained. This has confirmed applicability of the FLUENT to designing this type of reactors. The formation of a reaction zone in the center of the reactor insulated from the chamber face walls with a buffer gas was experimentally demonstrated and supported by numerical modeling. A decreased temperature of the near-wall gas mixture indicates that the reaction zone is also insulated from other reactor walls. Ethylene was shown to be an efficient converter of laser energy to heat power at ethane pyrolysis.

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