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EXPERIMENTAL INVESTIGATION OF HEAT TRANSFER AND SIMULATION OF REACTION TUBE FURNACES

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Results of an investigation of heat transfer in reaction tubes of furnaces for the pyrolysis and conversion of hydrocarbons are presented. Mathematical models of a furnace are developed, and generalized relationships between the diameter of the reaction coil, its length, the pyrogas temperature, and the time of contact are obtained. Optimum dimensions of reaction tubes and coils are determined.

The petroleum refining and chemical industries employ reaction tube furnaces for the processes of thermal cracking, pyrolysis, and catalytic conversion of hydrocarbons and many other processes. In view of the need for more efficient consumption of oil and natural gas, work is continuously under way on improvement of the designs of these apparatuses.

We carried out a series of experimental investigations of chemical transformations of hydrocarbons and heat transfer in both hollow coils and reaction tubes filled with a catalyst. The work was carried out on pilot and large-scale installations. The main element of the experimental facilities was a reaction tube heated by electric coils or immersed in a fluidized bed of a heat carrier. In the latter case the heater is a lined rectangular chamber divided by a partition into two zones supplied independently with a fuel mixture. The fuel gas and the air are mixed in the space under the grate, and combustion occurs directly in the fluidized bed. We measured the flow rates of raw material and pyrolysis products, the flow temperature along the entire length of the reactor, the tube wall and fluidized-bed temperatures, and other parameters and performed a chromatographic analysis of the raw material and the products. It was found that the presence of endothermic chemical reactions intensified heat transfer from the inner surface of the reaction tubes to the gas flow. The mechanism of intensification is explained as follows. Heat is transferred to the gas flow from the reactor wall and, consequently, there is a temperature gradient over the tube radius. The temperature gradient leads to a change in the reaction rate along the radius. Moreover, there is always a velocity gradient (at least in the wall layer), and the contact time is variable over the reactor radius. This leads to a change in the degree of conversion and to the appearance of a gradient of concentrations over the reactor radius. The flux of substances that transfer chemical energy causes an increase in the general heat flux. Consequently, the thermal conductivity coefficient of the reacting gas mixture is increased. This coefficient can be considered to be a sum of two components: molecular and reactive.

The overall reaction of the pyrolysis process is endothermic with an appreciable thermal effect. As a result, the heat capacity of the reacting mixture c_p' (the effective heat capacity) is much higher than that of an inert gas, since the heat supplied is spent not only on increasing the heat content of the mixture, but also on compensating the endothermal effect of the reactions. This causes deformation of the radical temperature profiles, which also leads to an increase in the heat flux.

Investigations of heat transfer carried out in reaction tubes showed that the coefficient of heat transfer to a reacting gas flow α' can be determined, with an accuracy sufficient for engineering calculations, from dimensionless equations of convective heat transfer upon substituting into them the effective Prandtl number Pr':

$$\mathrm{Nu}' = A\,\mathrm{Re}^{n}(\mathrm{Pr}')^{m}, \quad \mathrm{Pr}' = \mu c_p'/\lambda' .$$

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The following formula gives satisfactory results:

$$\alpha' = 0.026 w^{0.8} \rho^{0.8} \mu^{-0.4} (\lambda')^{0.6} (c_p')^{0.4} d^{-0.2}.$$

To create resource-saving installations, improved computational methods and mathematical models of furnaces were developed that involve problems of the kinetics of chemical reactions, internal and external heat transfer, and the efficiency of operation of a unit.

In simulating reaction tubes for the pyrolysis and conversion of hydrocarbons a two-parameter model was used, since due to the supply of heat and the chemical reactions there is a gradient of temperatures and concentrations along both the length and radius of the reactor. The processes occurring in the reactor are described by a system of partial differential equations:

$$\lambda_{ef} \left(\frac{\partial^2 t}{\partial r^2} + \frac{1}{r} \frac{\partial t}{\partial r} \right) - w(r) c_p' \frac{\partial t}{\partial l} - \omega^* \Delta H = 0,$$

$$D_{ef} \left(\frac{\partial^2 C_i}{\partial r^2} + \frac{1}{r} \frac{\partial t}{\partial r} \right) - w(r) \frac{\partial C_i}{\partial l} - \omega^* = 0.$$

For an externally heated cylindrical reactor with a fixed bed of a catalyst the boundary and initial conditions are

$$\text{when} \quad r = R \quad \lambda_{ef} \frac{\partial t}{\partial r} = -\alpha' (t_w - t), \quad \frac{\partial C_i}{\partial r} = 0;$$

$$\text{when} \quad r = 0 \quad \frac{\partial t}{\partial r} = 0, \quad \frac{\partial C_i}{\partial r} = 0;$$

$$\text{when} \quad l = 0 \quad C = C_0, \quad t = t_0, \quad w = w_0.$$

In the majority of cases, for industrial reactors it is possible to neglect the change in the flow velocity along the radius ($w(r) = \text{const}$). This simplifies the solution of the problem.

In hollow pyrolysis coils a gas flow moves with a high velocity (100–300 m/sec) and is highly turbulent. The wall boundary layer, which has a higher temperature than the flow core, occupies an insignificant fraction of the tube cross section. But there the reactions of destruction of hydrocarbons proceeed at a much higher rate, and this should be taken into consideration. For this purpose, we conventionally divide the coil into two reaction volumes: the flow core and the wall layer. The flow core can be described as a reactor of ideal displacement, and the wall layer as a reactor with radial transfer and parabolic distribution of velocities with the parabola axis located at the boundary between the layer and the flow core:

$$w(r) = w_f - w_f (r + \delta - R)^2 / \delta.$$

Consequently, we have the following boundary conditions:

$$\text{when} \quad r = R \quad \lambda_{ef} \frac{\partial t}{\partial r} = -\alpha' (t_w - t), \quad \frac{\partial C_i}{\partial r} = 0;$$

$$\text{when} \quad r = R - \delta \quad \frac{\partial t}{\partial r} = 0, \quad D_{ef} \frac{\partial C_i}{\partial r} = -\beta (C_i - C_{if}),$$

The simulation gives the distribution of temperatures and concentrations of the components of the raw material and the reaction products along the length and radius of the reaction tubes.

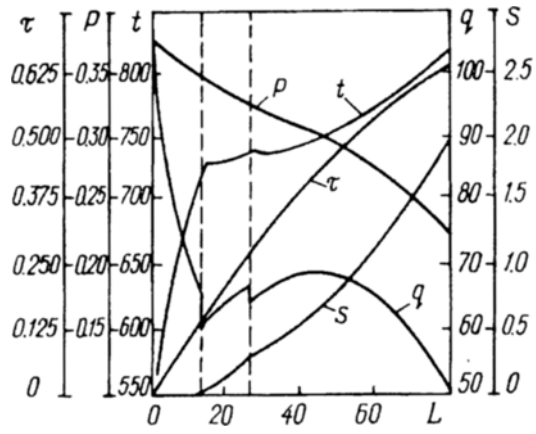


Fig. 1. Change in the parameters of the process of gasoline pyrolysis along the length of the coil. P, pressure, MPa; t, temperature, °C; τ, time of contact, sec; q, thermal stress, kW/m^2; S, degree of raw-material conversion; L, m.

For high-temperature tube furnaces to be optimally designed, the zone method of calculation of the combustion chamber is needed. It allows one to obtain a detailed picture of the distribution of heat fluxes and the temperature field in the radiation chamber. The basis of the zone method is that the internal space of the furnace is divided into a sufficiently large number of volumetric and surface zones (cellular model), and for each of these zones a heat balance equation is written in algebraic form:

$$Q_{rj} + Q_{cj} + Q_{tj} + Q_{sj} = 0 ,$$

where Q_r, Q_c, and Q_t are the resultant radiative, convective, and turbulent transfer of heat in zone j, respectively; Q_s are the heat sources (sinks) in the zone. Similar equations are written for all N zones. After substitution of the corresponding expressions for the heat transfer components, the system of heat balance equations is reduced to the form

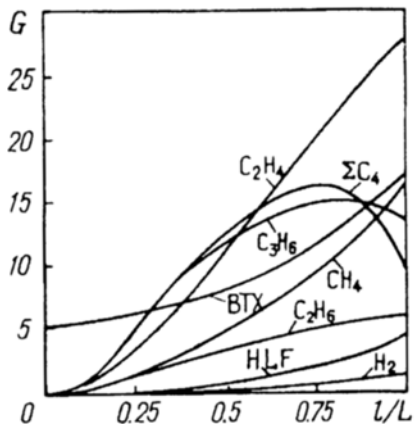
$$\sum_i^S A_{ij}T^4 + \sum_i^S B_{ij}T + \sum_i^S B_{jj}T + D\,(A_{ij}, B_{ij}, T_i, Q_s) = 0 ,$$

where A and B are the coefficients of radiative and convective transfer between the zones.

Radiative and convective heat fluxes from zones with prescribed temperatures are calculated and referred to the free term D. First, the angular coefficients of radiation between the zones, the heat emission along the length of the flame, the heat transfer coefficients, and other quantities were calculated. The solution of the system of equations gives the values of the temperatures averaged over the zones. Heat fluxes to the heating surfaces are determined by substituting the found values of temperatures into the equations for the zones of the heating surface.

On the basis of the calculation algorithms for the kinetics of chemical reactions, reaction coils, fuel combustion, and radiative and convective chambers, a mathematical model of the furnace as a whole was developed. It is based on the block principle and operates as follows. First, the technological element of the furnace is simulated, i.e., the radiation tube or the coil, and, using the data obtained, the radiation chamber is calculated. The results are transmitted to the block for calculation of the convective zone of the furnace; the temperature determined here is compared with that adopted for the calculation of the coil. If their values do not coincide, then the value of the temperature obtained is introduced, and a new cycle of calculation is started until the values of the parameters coincide within the accuracy prescribed. In this way the material and heat balances for the entire unit are interrelated. Next, the specific discharge of the fuel, the energy and exergy efficiencies, and other technical-and-economic indices of furnace operation are calculated.

We simulated the operation of different-purpose furnaces. As an example, Fig. 1 presents some results of simulation of a gasoline pyrolysis furnace involving a complex coil, in which four tubes are united into two and



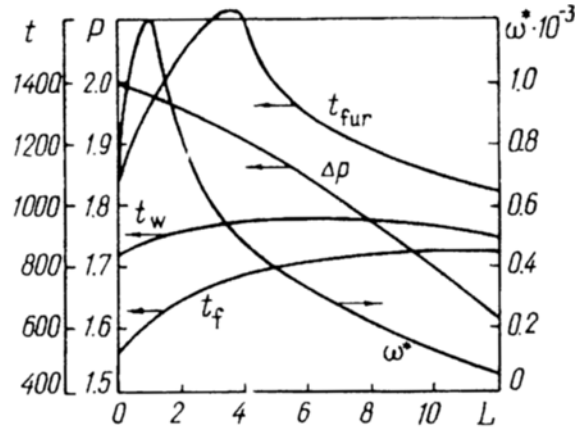


Fig. 2. Distribution of gasoline pyrolysis products G, wt.%, along the coil length; BTX, benzene-toluene-xylene fraction; HLF, heavy liquid fuel; l/L, relative length.

Fig. 3. Change in the parameters of the process of vapor conversion of natural gas along the length of the reaction tube L, m. t_{fur}, t_w, t_f, temperatures of the furnace, the reactor wall, and the flow, °C; Δp, drop in pressure, MPa; ω^*, mole/m^2·sec.

then into one tube of large diameter. The dashed lines show the places of junction of the tubes. When a gas flow moves along the reaction coil, the temperature rises, but when it attains 700°C its growth slows down, since a considerable portion of the heat supplied begins to be absorbed by reactions of raw-material decomposition. As the pressure falls and the molecular mass of the mixture decreases, the flow velocity increases, and the slope of the contact time curve becomes smaller. The degree of raw-material decomposition increases sharply with increase in temperature. The yield of pyrolysis products (Fig. 2) depends mainly on the stability of the process or degree of raw-material decomposition, which increases along the coil length.

As a result of simulation of furnaces, relationships were obtained between the temperature in the reaction chamber, temperature of the tube wall, outlet temperature of the pyrogas and contact time, coil diameter, and its length at fixed values of the degree of raw-material conversion or the stability of the process. A decrease in the diameter leads to a substantial decrease in the tube wall temperature, but here the efficiency of the coil is decreased, and a large number of tubes are required to ensure the needed power of the furnace. The load on the coil is limited by the temperature of the tube wall and the flow velocity from the conditions of abrasive wear.

With increase in the prescribed degree of raw-material conversion or stability of the process, the consumption of heat for pyrolysis is increased, as is the fuel consumption. As a result, in the convective zone of the furnace a larger quantity of heat is transferred to the raw material, and the temperature of the raw material at passage into the radiative zone is increased. The temperature of the waste gases and the efficiency of the furnace hardly change in the operational range of the parameters.

We simulated a furnace for catalytic steam and steam-carbon dioxide conversion of natural gas. As a result, we obtained values of the flow parameters, thermal characteristics, and composition of the products of conversion along the reaction tube length (Fig. 3). The temperature of the furnace rises rapidly due to intense heat generation by the flame and then falls, since the heat is absorbed by the reaction tubes. The observed reaction rate first also increases due to the rapid rise in the temperature but then falls due to the decrease in the concentration of reagents (CH_4 and H_2O).

Simulation of conversion furnaces showed that an increase in loading with raw material entails an increase in the temperature of the converted gas, since it is necessary to increase the reaction rate and maintain the prescribed degree of conversion of methane. The temperature of the tube wall is increased correspondingly in spite of the increase in the heat transfer coefficient.

At the present time, highly efficient methanol and ammonia installations are being built in which the overall number of tubes in the furnaces will attain 1000. This lowers the reliability of operation of the furnaces and the entire unit. Therefore it is necessary to use tubes of the maximum possible capacity. As the diameter increases, the conditions of heat transfer sharply deteriorate, resulting in a reduction in the maximum allowable mass velocity. Consequently, there is an optimum diameter, exceeding which does not lead to an increase in the capacity of the tube, i.e., there is a capacity extremum. As the tube length is increased, the optimum diameter increases. For a diameter of the reaction tube of 70–75 mm the optimum length of the working zone of the tube is 14 m, and for a diameter of 60–65 mm, it is 10 m. Optimization of the geometric dimensions of the reaction tubes and other elements of the furnace leads to substantial economizing in refractory alloys.

The results of the calculations of the processes and elements of a furnace are transmitted to the computational block where the main technical-and-economic indices of the furnace are determined: consumption of fuel, steam, and electrical energy, the overall energy expenditure, and the thermal and energy efficiency. A thermodynamic analysis is also carried out, and the energy efficiency of the unit is determined.

The main exergy losses are attributable to the following: irreversibility of chemical conversions, including combustion processes; nonequilibrium heat exchange between combustion products and flows in the coils; a drop in pressure in the coils; mixing of product flows and dilution of them with steam; radiation of heat into the ambient and its escape with the flue gases.

The exergy efficiency of a furnace depends on both the internal and external losses and may comprise 25–70%, but in the majority of modern furnaces it usually lies within the range of 45–65%.

As a result of experimental investigations and simulation of facilities for pyrolysis and conversion of hydrocarbons, optimum diameters and lengths of reaction tubes and coils were determined, resulting in economizing in refractory alloys. The determined optimum profiles of heat fluxes ensure an increase in the yield of products by 4–6%. Use of the methods of mathematical simulation makes it possible to create complex technological installations with optimum raw-material and energy expenditures and to reduce substantially the consumption of petroleum fractions and natural gas in chemical production.

NOTATION

c_p, heat capacity; λ, thermal conductivity; ρ, density; μ, coefficient of viscosity; w, rate; d, diameter of the reactor; α, coefficient of heat transfer; H, heat of the chemical reactions; t, temperature; r, radius; l, current length; L, length of the reactor; ω^*, observed rate of the chemical reactions; C, concentration; D, diffusion coefficient; δ, boundary layer thickness; β, mass transfer coefficient; Q, quantity of heat. Subscripts: ef, effective value; f, flow; w, wall.