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# STUDYING HEAT EXCHANGE OF DISPERSION-DROP GAS–LIQUID FLOW IN GRANULAR LAYER

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**Introduction**. In some processes of chemical technology, e.g., in catalytic decomposition of monopropellants, there are used catalytic layers heated to high temperatures through which dispersion gas–liquid flow passes. Evaporation of drops, whose vapors react on a catalyst, heating it to high temperatures, can become a limiting stage of the process. The quantity of the evaporated liquid is directly proportional to the quantity of heat, transmitted from the catalyst to the liquid either through gas phase or directly to drops. This process is not studied yet and the dependences of heat exchange on the basic parameters of the process are not known, namely, flow intensity of gas and liquid, gas and catalyst temperature, and dimensions of a grain and drops. Investigation procedure has not been devised.

**Investigation Procedure**. The main difficulty of the experiment was to supply a great heating rate to the layer. The value of the heating rate must not depend on the process of evaporation, in order to keep the analysis of the experiment from becoming too involved.

Electric heating of the layer would not do in this case. First, it is technically difficult to realize; second, it is impossible in principle to provide uniform heating of the granular layer, since maximum electric heating occurs at the points of grain contact.

The gas carrying drops and preheated to high temperatures can be used for heating the layer. We tested this procedure but rejected it because at the moment when drops are injected into the flow and when they pass the distance from the injector to the layer, heat exchange between the drops and heated gas flow takes place, which complicates the experiment and yields great errors. In addition, due to low heat capacity of a gas as compared to a heat of evaporation of a liquid, this method can be used only for very small ratios of the liquid–gas flow rates.

In our procedure for the heating of the layer we used oxidation reaction of hydrogen by atmospheric oxygen on the catalyst layer itself, which proceeds in external diffusion conditions. Heat release on the layer could be varied independently, that is, by changing hydrogen concentration at the inlet to the reactor or by changing the flow rate of hydrogen–air mixture with a constant concentration. Using inert liquid (water) which was supplied by a pneumatic injector to the layer in the form of drops practically eliminated the influence of heat exchange and evaporation on the heat release process, and where this influence was appreciable, it was easily considered according to a measured amount of the reacted hydrogen.

"Thin" layer of the catalyst was located in a vertical quartz tube with internal diameter 23 mm. The accompanying gas–liquid flow was supplied to the layer from top to bottom. The height of the layer was selected in such a way that on the one hand not all the liquid evaporates and, on the other hand, evaporation would be sufficiently high for measuring. Within the limits of a grain diameter variation 0.7-3.22 mm the height of the layer was selected within the limits 2.5-10 mm, respectively.

For the layer we used a catalyst of two types, namely, granules of aluminum oxide and pore-metallic grains. In both cases, coated platinum was an active component. Granules from oxide were close to spheric ones with an average diameter 3.2 and 2.5 mm, a mixture of these crushed grains had diameters 1.25 and 0.75 mm, and pore-metallic grains head an average diameter 2.5 mm.

If the initial concentration of hydrogen was changed within 5-10% (volume percent) and specific consumption of gas was 1.8-5.3 $m^3/m^2$ (here and below gas flow rate is given in normal conditions), then layer temperature varied within 400-800°C. The injector was installed along the tube's axis at such distance that at given flow rates of gas and liquid the diameter of spray torch corresponds to the diameter of the tube. By changing the air flow rate through the injector we varied an average size of drops within 50-200 $\mu$.

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Volume surface diameter of a drop was determined by the method of printing on a thick layer of carbon black, coated on a glass plate:

$$d = (\Sigma d_i^3 n_i)/(\Sigma d_i^2 n_i)$$

($n_i$ is the number of drops with the diameter $d_i \pm 5\ \mu$, measured and calculated with the help of a tool microscope on the plate after it was exposed to a spray torch). With this diameter the surface of drops is correctly determined for a given volume of liquid [1].

Liquid and gas flow rates, temperatures of the catalyst and gas flow, and hydrogen concentration at the layer outlet were measured in stationary conditions. The gas temperature was measured by a comb of six thermocouples at a distance of 10 mm from the lower surface of the layer. Preliminary experiments with thermocouples protected from drops and thermocouples with "open heads" have not revealed any significant difference in their readings. Hydrogen concentration at the layer inlet was determined by the flow rate of hydrogen and air and at the outlet, by a chromatograph.

The amount of heat q, transmitted to water, was found from the balance equation

$$Q_h y H = c_p Q(T - T_0) + q_1 + q, \tag{1}$$

where $Q_h$, Q is the flow rate of hydrogen and vapor–gas mixture at the layer outlet; y is the degree of hydrogen conversion; H is the thermal effect of the reaction of water formation; $c_p$ is an average heat capacity of the gaseous mixture upon the change of temperature from $T_0$ to T; and $q_1 = \alpha_1(T - T_0)$ are the heat losses into the environment.

In order to reduce systematic errors, the amount of heat transmitted to water was determined from two measurements in stationary conditions: with water supplied to the layer and without water (with other parameters being unchanged). Heat transfer coefficient with the environment $\alpha$ can be found from the equation corresponding to the case without water:

$$Q_h y_1 H = c_{p1} Q_1 (T_1 - T_0) + \alpha_1 (T_1 - T_0). \tag{2}$$

Its value in the experiment is 0.07 W/deg. The quantities $c_{p1}Q_1$, $c_pQ$ are little different from each other due to an insignificant difference in the degree of conversion and a relatively small change of gas temperature at the outlet. With due regard to these assumptions from (1) and (2) we obtain

$$q = (c_p Q + \alpha)(T_1 - T) - Q_h H(y_1 - y). \tag{3}$$

To estimate the heat transfer efficiency to the drops in a unit volume of the layer, we calculated the value of the volume heat transfer coefficient

$$k = q/(V\ T),\ \mathrm{W/m^3/°C} \tag{4}$$

(V is the volume of the layer and T is the gas temperature at the layer outlet).

The amount of evaporated water was calculated from the formula

$$G_1 = q/(h(T) - h(T_0))$$

(h(T) is water enthalpy for the corresponding temperatures). In this calculation the heat spent on heating of the remaining liquid was neglected because the temperature of drops at the outlet is unknown. If we assumed that the remaining water was heated to 100°C, then the error would be 20%.

**Discussion.** The results of experiments with a catalyst are given in Table 1, where $T_1$ and $T_2$ is the catalyst temperature at the layer inlet and outlet. In the range of the studied parameters and within the error of the experiment the value of k depends only slightly on the drop dimensions and gas velocity. For pore-metallic grains k is twice that for ceramic grains, all other things being equal.

The dependence of k on $d_3$ is given in Fig. 1. Here for the ceramic catalyst for d = 0.1-0.2 mm, Q = 760-1100 $\mathrm{cm^3/s}$ points 1 and lines I correspond to G = 0.02 $\mathrm{cm^3/s}$, 2 and II correspond to G = 0.04 $\mathrm{cm^3/s}$, lines I, II correspond to the

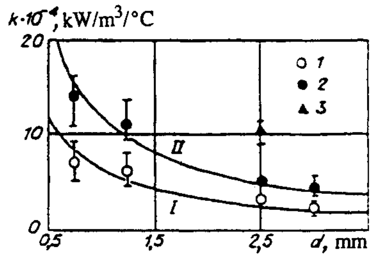

Fig. 1

| $Q$ | $Q_h$, cm$^3$/sec | $T_1$ | $T_2$ | $T$ | $G\ 10^2$, cm$^3$/sec | $d$, μm | $G_1\cdot10^2$, cm$^3$/sec | $k\cdot10^{-4}$, W/m$^3$/°C |
|---|---|---|---|---|---|---|---|---|
| | | °C | | | | | | |
| 990 | 90 | 744 | 723 | 550 | 0 | — | — | — |
| 990 | 90 | 605 | 605 | 473 | 4,6 | 57 | 2 | 7,5 |
| 1323 | 113 | 546 | 547 | 451 | 4,9 | 57 | 2,9 | 10,9 |
| 2190 | 190 | 572 | 583 | 462 | 4,9 | 57 | 2,8 | 10.1 |
| 2190 | 190 | 627 | 627 | 479 | 4,9 | 100 | 2,7 | 9,5 |
| 2190 | 190 | — | — | 462 | 4,9 | 150 | 3 | 11 |

calculation by Eq. (5); for pore-metallic catalyst for d = 0.06-0.15 mm, Q = 990-2200 cm$^3$/s points 3 correspond to G = 0.049 cm$^3$/s.

A considerable dependence of the volume heat transfer coefficient on the liquid flow rate G and the catalyst grain size $d_3$ is seen from the figure. The dependence can be expressed as

$$k = AG/d_3, \tag{5}$$

where A is the proportionality factor, selected for k = $13\cdot10^4$ W/m$^3$/°C, G = 0.04 cm$^3$/s, $d_3$ = 1 mm.

To find out the heat exchange mechanism of the drops in the layer let us calculate the coefficient k for two cases: (1) the drops move through the layer with a velocity of gas; (2) the drops are decelerated during collisions with grains, then are accelerated by gas.

For the first case the heat transfer coefficient of a drop with the flow is $\alpha = \mathrm{Nu}\lambda/d$ (Nu = 2, λ is the gas thermal conductivity). The number of drops inflowing the layer is $n = 6G/(\pi d^3)$. The duration of their stay in the layer volume is $\tau = V\varepsilon/Q_3$ ($Q_3 = Q(T + 273)/273$). The amount of heat transmitted to the drops is $q = \pi d^2\alpha n\tau(T - T_0)$. Neglecting $T_0$ as compared to T and using (4), we obtain

$$k = 12\lambda\varepsilon G/(d^2Q_3). \tag{6}$$

Calculating this quantity with the parameters corresponding to the experiment (T = 500°C, d = 0.1 mm, Q = $10^3$ cm$^3$/s, G = $4\cdot10^{-2}$ cm$^3$/s, ε = 0.4 is the layer porosity) yields the value of k = 400 W/m$^3$/°C, which is 260 times less than that found in the experiment for a ceramic grain with a diameter of 1.25 mm, and 128 times less than with a diameter 3.2 mm. For a pore-metalic grain this difference will be still more.

A comparison of experimentally obtained dependence (5) with theoretical (6) points to a qualitative and quantitative nonconformity. To provide such a great heat exchange between the drops and gas, it is necessary that the drops were crushed or their velocity of motion through the layer was much lower than the gas velocity. Measurements of the drop dimensions have shown that their average diameter changes only little and almost corresponds to a reduction of drop due to evaporation (average diameter changed from 57 to 42 μ). In addition, Weber's number calculated for the conditions of the experiment is less than 1, which indicates that the drops are stable to crushing.

Let us estimate the second case for $d_3 = l$ mm and with the following assumptions: the number of collisions of a drop in a unit volume of the layer is $n_1 = (6(1 - \varepsilon)/\pi)^{1/3}/d_3$, the distance between the collisions on a unit length of the layer is $l = 1/n_1$, time $t_1$ for which a drop flies the distance $x = l$ we find from the equation of motion of a drop:

$$m\delta^2x/\delta t^2 = F \text{ при } t = 0,\ x = 0,\ \delta x/\delta t = 0.$$

The force acting on the drop $F = c\rho(u - w)^2\pi d^2/8$ ($c = f(\mathrm{Re})$, $\mathrm{Re} = (u - w)d/\nu$, u, ρ, ν is the velocity, density, kinematic viscosity of a gas in the layer). A drop's mass $m = \rho_2\pi d^3/6$ ($\rho_2$ is the liquid density). Residence time of a drop in a unit volume of the layer is $\tau = t_1n_1$.

Analyzing the solution for the above-mentioned parameters of the layer and flow, we obtain u >> w, $t_1 \approx (2lm/F)^{0.5}$ [2, pp. 25, 149]. The volume heat transfer coefficient of a drop with gas in the layer is

$$k = \tau n \alpha \pi d^2 \approx 1{,}7\lambda u_2 [\rho_2 (1 - \varepsilon)^{1/3}/(d d_3 \rho)]^{0.5}/\nu \quad (7)$$

($u_2$ is the specific flow rate of the liquid).

Equation (7) corresponds to the experiment quantitatively and qualitatively better than (6). The calculations made according to this model show that for the above-given parameters of the layer and flow a stationary drop would be subjected to acceleration from gas of the order of 1000 m/s$^2$. Here, however, the heat transfer coefficient is only doubled and the residence time in the layer is increased by 20 times as compared to the drop trapped into the flow. These estimates yield an increase in the volume coefficient by 40 times (($k = 1.6 \cdot 10^4$ W/m$^3$/°C), which is obviously insufficient to correspond to the experiment, where $k = 10.5 \cdot 10^4$ W/m$^3$/°C.

Probably the main heat exchange mechanism of a drop in a granular layer is the mechanism of interaction between drops and heated surface of a catalyst. In this case the volume heat transfer coefficient k must be proportional to: a dimensionless quantity $P \sim d/d_3$, i.e., a probability of collision of a drop in the layer, which is proportional to the surface of a unit volume of the layer $1/d_3$ and the drop's diameter d; $n \sim u_2/d^3$ the number of drops inflowing a unit volume; $s \sim d^2$ the surface of the drop, i.e., $k = Pns \sim u_2/d_3$, which correspond to Eq. (5), obtained experimentally.

The last model is also supported by an increase of the volume heat transfer coefficient with heat capacity and thermal conductivity of the catalyst grain, found experimentally. These parameters are greater for pore-metallic grains than for ceramic ones. When a drop interacts with a surface, the amount of the transmitted heat must depend on thermal properties of the material of this surface, i.e., on the catalyst grain, as it was in our experiments.

**Conclusions.** Experimental procedure has been developed for physical modeling of monopropellant decomposition in a catalytic packet upon limiting stage of the process, i.e., during evaporation of a liquid in drop conditions. Heat exchange of liquid drops in a catalyst layer heated to high temperatures has been analyzed. Experimental dependence of a volume heat transfer coefficient on grain diameter, liquid flow rate and catalyst material has been obtained. It has been shown that within parameter variations this coefficient is practically independent of the gas velocity and drop diameter. Evaporation mechanism of drops in a heated granular layer has been discussed and carried out.

For a more comprehensive examination of the interaction mechanism between the drops and the catalyst layer, a further experimental investigation is necessary in a wider range of change of the basic parameters of the process and use of mathematical modeling in analyzing experimental data.

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