HEAT AND MASS TRANSFER IN CHEMICALLY REACTING MEDIA WITH DECREASE IN PARTIAL PRESSURES OF VAPOR

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We present a mathematical model of heat and mass transfer in a chemically reacting medium—multicomponent vapor-gas mixture system with decrease in partial pressures of vapor-gas components in a free reactor volume.

Technological processes occurring with a pressure decrease are implemented in hermetically sealed apparatuses and are characterized by minimum ejections of a vaporable liquid into the surrounding medium [1]. By the degree of change in the mechanical equilibrium in systems with a different phase state of the media it is possible to separate two groups of processes: processes occurring with decrease in the residual (total) pressure of a vapor-gas medium over the surface of a moist material or solution [2, 3] and also processes occurring with decrease in the excess (partial) pressure. Among the latter is the chemical interaction of the reagents in liquid multiphase media which is accompanied by liberation of a great amount of gases and reaction heat, resulting in an increase in the temperature and intense evaporation of components of a reaction mixture.

A mathematical description of heat and mass transfer in chemically reacting media with decrease in the vapor pressure is based on the Umov equation [4], which represents a generalized model of potential transfer

$$\frac{\partial \varphi}{\partial \tau} + \operatorname{div}(\varphi w) = -\operatorname{div} q + \gamma, \tag{1}$$

where the first term on the left-hand side is the intensity of change in the transfer potential φ, while the second term is the macroscopic potential motion; the first term on the right-hand side of Eq. (1) is the potential-transfer flux q, while the second term is the potential source (sink) γ.

In order to take into account all the factors affecting the heat and mass transfer, we consider a vapor-gas mixture—multicomponent liquid system with chemical interaction of the reagents in the latter. As the assumption that concretizes Eq. (1), we adopted the model of ideal mixing of components in coexisting phases. This circumstance is attributable to the decrease in the partial pressures of the components of the mixture that decreases the resistance to the transfer of vapor-gas components into it [5], and also to the intense volumetric interaction of the reaction-mixture reagents and high degree of turbulence of the liquid phase.

Vapor-Gas Phase. The difference in the partial pressures of the vapor of the moisture removed over the solution surface and in the surrounding medium [6] is the driving force of all the processes occurring with decrease in the medium pressure

$$j_{i(j)} = \frac{\beta_{i(j)} \, \mu_{i(j)}}{R^* T_{\text{liq.s}}} (p_{i(j)\text{s}} - p_{i(j)}). \tag{2}$$

The equilibrium values for the partial pressures of the vapor in close proximity to the reaction-mixture surface $p_{i(j)\text{s}}$ are established instantly in conformity with the liquid-phase temperature and concentration of the reaction products.

The partial pressure of the component in the surrounding medium $p_{i(j)}$ is determined from formula (1), which is represented by a system of differential equations of mass and energy transfer [7] with the volume sources (sinks) of heat and mass. For the case of ideal mixing of the components in the vapor-gas phase [8]

$$\frac{d\rho_{i(j)}}{d\tau} = \rho_{i(j)}\left(\frac{d\bar{y}_{i(j)\text{inl}}}{d\tau} - \frac{d\bar{y}_{i(j)}}{d\tau}\right), \tag{3}$$

$$\frac{d\rho_g}{d\tau} = 0, \tag{4}$$

$$c_{\mu\,\text{mix}}\,\rho_{\text{mix}}\frac{dT}{d\tau} = \left[\alpha\,(T_{\text{liq.s}} - T) + T_{\text{liq.s}}\left(\sum_{i=1}^{n} j_i\,c_i + \sum_{j=1}^{m} j_j\,c_j\right)\right]\frac{\mu_{\text{mix}}}{R_{\text{v.g}}} - c_{\mu\,\text{v.g}}\,\rho_{\text{v.g}}T\times$$

$$\times \sum_{i=1}^{n}\frac{d\bar{y}_i}{d\tau} + \sum_{j=1}^{m}\frac{d\bar{y}_j}{d\tau}. \tag{5}$$

The left-hand side of Eq. (3) characterizes the rate of change in the mass of the $i(j)$-th component of the vapor-gas phase per unit free volume of the apparatus; the first term on the right-hand side determines the rate of the supply of the $i(j)$-th component mass into the vapor-gas phase due to evaporation and chemical reaction; the second term on the right-hand side determines the rate of the component removal into a system of withdrawal of the vapor-gas mixture.

As the source of rarefaction for the processes occurring with decrease in the partial pressures of vapor and gases, in practice use is made of liquid-jet ejectors [9]. A liquid jet, issuing from a nozzle into a receiving chamber, entrains the vapor-gas mixture from a reactor; here the working liquid is an absorbent relative to the liberated gases. Because of the high intensity of the heat and mass transfer, absorption and condensation of the vapor-gas mixture are provided on the jet.

The index of the efficient operation of the liquid-jet ejectors is the volumetric coefficient of injection [9]

$$U_{\text{v.max}} = \frac{Q_{\text{v.g}}}{Q_{\text{w}}} = \frac{0.35\,(p_{\text{w}} - p_{\text{m}})}{0.37p_{\text{m}} - p_{\text{sat}}}, \tag{6}$$

which depends on the structural parameters of the equipment and the temperature of the working liquid.

In the short-term starting operation period, the liquid-jet ejector provides the maximum possible rarefaction in the hermetic chamber. Hereafter, according to Eq. (4), the composition of the inert gas (air) in the chamber remains constant, and the mixture of vapor and gases, liberated due to the reaction, is the injected medium.

In the equation of energy transfer (5) the left-hand side is the rate of change in the heat content of the multicomponent vapor-gas mixture; the right-hand side characterizes the heat exchange of the vapor-gas mixture with the surface of the coexisting phase, the heat inflow per unit free volume of the apparatus with the vapor of the vaporizable liquid and gas components formed due to the chemical reaction, and also the heat removal with the vapor-gas mixture into the system of its withdrawal.

We determine the extensive quantities entering into Eqs. (4) and (6) by the rule of additivity [10]. The densities of the vapor-gas mixture contained in the free apparatus volume and removed into the mixing chamber of the ejector will respectively be written by the equations

$$\rho_{mix} = \frac{1}{R^* T}\left(\mu_g p_g + \sum_{i=1}^{n} \mu_i p_i + \sum_{j=1}^{m} \mu_j p_j\right),$$ (7)

$$\rho_{v.g} = \frac{1}{R^* T}\left(\sum_{i=1}^{n} \mu_i p_i + \sum_{j=1}^{m} \mu_j p_j\right);$$ (8)

the molar heat capacities, by the equations

$$c_{\mu\,v.g} = \frac{c_{\mu g} p_g + \sum_{i=1}^{n} c_{\mu i} p_i + \sum_{j=1}^{m} c_{\mu j} p_j}{p_g + \sum_{i=1}^{n} p_i + \sum_{j=1}^{m} p_j},$$ (9)

$$c_{\mu\,v.g} = \frac{\sum_{i=1}^{n} c_{\mu i} p_i + \sum_{j=1}^{m} c_{\mu j} p_j}{p_g + \sum_{i=1}^{n} p_i + \sum_{j=1}^{m} p_j};$$ (10)

and the molecular mass, by

$$\mu_{mix} = \frac{\mu_g p_g + \sum_{i=1}^{n} \mu_i p_i + \sum_{j=1}^{m} \mu_j p_j}{p_g + \sum_{i=1}^{n} p_i + \sum_{j=1}^{m} p_j}.$$ (11)

After the reduction to the free volume of the apparatus V_f, in which the vapor-gas mixture is located, substitutions in the form of relations (7)-(11), the Mendeleev–Clapeyron equation [10], and also of the equations

$$V_f \rho_{i(j)} \frac{d \bar{y}_{i(j)inl}}{d\tau} = j_{i(j)} F,$$ (12)

$$V_f\left(\sum_{i=1}^{n} \frac{d \bar{y}_i}{d\tau} + \sum_{j=1}^{m} \frac{d \bar{y}_j}{d\tau}\right) = Q_{inl},$$ (13)

and differentiation and corresponding transformations, the system of equations (4)-(6) can be represented as

$$\frac{dp_{i(j)}}{d\tau} = \frac{FR^* T}{V_f \mu_{i(j)}} j_{i(j)} - p_{i(j)}\left(\frac{Q_{v.g}}{V_f} - \frac{1}{T}\frac{dT}{d\tau}\right),$$ (14)

$$p_g = \text{const},\tag{15}$$

$$\frac{dT}{d\tau} = \left\{ \frac{\left[\alpha(T_{\text{liq}} - T) + T_{\text{liq}}\left(\sum_{i=1}^{n} j_i\, c_i + \sum_{j=1}^{m} j_j\, c_j\right)\right] FR^*}{c_{\mu g}\, p_g + \sum_{i=1}^{n} c_{\mu i}\, p_i + \sum_{j=1}^{m} c_{\mu j}\, p_j} - \frac{\sum_{i=1}^{n} c_{\mu i}\, p_i + \sum_{j=1}^{m} c_{\mu j}\, p_j}{\mu_g\, p_g + \sum_{i=1}^{n} \mu_i\, p_i + \sum_{j=1}^{m} \mu_j\, p_j} \times \right.$$

$$\left. \times \frac{\sum_{i=1}^{n} \mu_i\, p_i + \sum_{j=1}^{m} \mu_j\, p_j}{c_{\mu g}\, p_g + \sum_{i=1}^{n} c_{\mu i}\, p_i + \sum_{j=1}^{m} c_{\mu j}\, p_j}\, Q_{\text{v.g}} \right\} \frac{T}{V_f}.\tag{16}$$

Liquid Phase. For the liquid phase the differential equations of energy and mass transfer with allowance for the assumptions taken have the form

$$c_{\text{liq}}\, \rho_{\text{liq}}\, \frac{dT_{\text{liq}}}{d\tau} = \left[\alpha\,(T - T_{\text{liq}}) - T_{\text{liq}}\left(\sum_{i=1}^{n} j_i\, c_i + \sum_{j=1}^{m} j_j\, c_j\right) - \sum_{i=1}^{n} j_i\, r_i\right]\frac{1}{R_{\text{v.liq}}} +$$

$$+ \rho_{\text{liq}} q_{\text{ch.r}} \sum_{i=1}^{n} \frac{d\bar{x}_{i\text{ch.r}}}{d\tau},\tag{17}$$

$$\frac{d\rho_{\text{liq}i}}{d\tau} = -\rho_{\text{liq}i}\left(\frac{d\bar{x}_i}{d\tau} + \frac{d\bar{x}_{i\text{ch.r}}}{d\tau}\right).\tag{18}$$

The left-hand side of Eq. (17) determines the heat content of the liquid mixture; the first term of the right-hand side characterizes the heat exchange of the vapor-gas phase with the liquid-mixture surface; the second term characterizes the heat removal with the vapor-gas mixture into the free apparatus volume; the third term, the heat consumption caused by a phase transition of the reagents of the liquid phase; the fourth term characterizes the heat supply due to the exothermal reaction.

In Eq. (18), the left-hand side is the rate of change in the mass of the i-th component per unit volume of the liquid phase; the first term on the right-hand side is the rate of removal of the liquid-reagent mass into the vapor-gas phase due to evaporation; the second term is the rate of chemical transformation of the component. We present the density and mass heat capacity of the liquid mixture by the relations [10]

$$\rho_{\text{liq}} = \sum_{i=1}^{n} \rho_{\text{liq}i}\, \frac{\bar{x}_i}{\mu_i},\tag{19}$$

$$c_{\text{liq}} = \sum_{i=1}^{n} \bar{x}_i\, c_i.\tag{20}$$

The intensities of change in the mass of the liquid-mixture reagents and of the chemical-interaction products will be calculated on the basis of the kinetic equation of reaction [11]:

for homogeneous systems

$$\pm \frac{dC_{i(j)}}{d\tau} = k \prod_{i=1}^{n} C_i^{\chi_i} ; \tag{21}$$

for heterogeneous systems

$$\pm \frac{dC_{i(j)}}{d\tau} = k'S \prod_{i=1}^{n} C_i^{\chi_i} , \tag{22}$$

in Eqs. (21) and (22) the minus sign refers to the reagents, while the plus sign refers to the reaction products. We express the volume concentrations of the components in terms of the mass fractions [8]

$$C_i = \frac{\rho_{\text{liq}} \overline{x}_i}{\mu_i} \tag{23}$$

or

$$\frac{dC_i}{d\tau} = \frac{\rho_{\text{liq}}}{\mu_i} \frac{d \overline{x}_i}{d\tau} . \tag{24}$$

Substitution of relation (24) into Eqs. (21) and (22) gives

$$\frac{d \overline{x}_{\text{ich.r}}}{d\tau} = \rho_{\text{liq}}^{(\chi_i - 1)} \mu_i^{(1 - \chi_i)} k \prod_{i=1}^{n} \overline{x}_i^{\chi_i} , \tag{25}$$

$$\frac{d \overline{x}_{\text{ich.r}}}{d\tau} = \rho_{\text{liq}}^{(\chi_i - 1)} \mu_i^{(1 - \chi_i)} k'S \prod_{i=1}^{n} \overline{x}_i^{\chi_i} . \tag{26}$$

After the reduction to the volume V_{liq} in which the liquid mixture is contained and after transformations, Eq. (17) takes the form

$$\frac{dT_{\text{liq}}}{d\tau} = \left[\frac{\alpha (T - T_{\text{liq}}) - T_{\text{liq}} \left(\sum_{i=1}^{n} j_i c_i + \sum_{j=1}^{m} j_j c_j \right) - \sum_{i=1}^{n} j_i r_i}{c_{\text{liq}} \rho_{\text{liq}}} \right] \frac{F}{V_{\text{liq}}} +$$

$$+ \frac{q_{\text{ch.r}} \left(\sum_{i=1}^{n} \rho_{\text{liq}}^{\chi_i} \mu_i^{(1 - \chi_i)} k \prod_{i=1}^{n} \overline{x}_i^{\chi_i} \right)}{c_{\text{liq}} \rho_{\text{liq}}} . \tag{27}$$

The thermal effect of the chemical reaction $q_{\text{ch.r}}$ is calculated by the Hess law [11] depending on the nature and state of the initial substances.

The rate of the removal of the liquid-reagent mass into the vapor-gas phase due to the evaporation can be determined from the Raoult law [10], which with account for the Antoine equation [12] will be written as

$$p_i = \frac{\overline{x_i}}{\mu_i} \exp\left(A_i - \frac{B_i}{T_{\text{liq}}}\right)$$

(28)

or as

$$\frac{d\overline{x_i}}{d\tau} = \frac{\dfrac{dp_i}{d\tau} - \dfrac{\overline{x_i}B_i}{\mu_i T_{\text{liq}}^2}\dfrac{dT_{\text{liq}}}{d\tau}\exp\left(A_i - \dfrac{B_i}{T_{\text{liq}}}\right)}{\exp\left(A_i - \dfrac{B_i}{T_{\text{liq}}}\right)}\mu_i .$$

(29)

By substituting relations (25) and (29) into Eq. (18) we obtain an equation which determines the change in the mass of the i-th component of the liquid mixture:

$$\frac{d\rho_{\text{liq}i}}{d\tau} = -\rho_{\text{liq}i}\,\mu_i \left[\frac{\dfrac{dp_i}{d\tau} - \dfrac{\overline{x_i}B_i}{\mu_i T_{\text{liq}}^2}\dfrac{dT_{\text{liq}}}{d\tau}\exp\left(A_i - \dfrac{B_i}{T_{\text{liq}}}\right)}{\exp\left(A_i - \dfrac{B_i}{T_{\text{liq}}}\right)} + \frac{\rho_{\text{liq}i}^{(\chi_i - 1)}}{\mu_i^{\chi_i}} k \prod_{i=1}^{n}\overline{x_i}^{\chi_i}\right].$$

(30)

Changes in the temperature of the liquid mixture and in the mass of its component reagents will be determined for the heterogeneous systems from Eqs. (27) and (30) with allowance for the interface considered from Eq. (22).

The constant of the reaction rate k that characterizes the occurrence of the process at a microlevel depends on the nature of the reacting substances and temperature. The dependence of the reaction constant on the temperature is usually represented in the form of the Arrhenius law [13]

$$k = k_0 \exp\left(-\frac{E_{\text{act}}}{R^* T_{\text{liq}}}\right).$$

(31)

The preexponential factor k_0 is determined by a numerical method according to the recommendations given in [13], while the activation energy E_{act} has numerical values lying within the limits of 40–400 kJ/kmol [14].

The solution of the system of equations describing a change in process parameters was carried out for the vapor-gas phase (14)-(16) and liquid mixture (27), (30), and (31) by a numerical method with the following conditions:

boundary conditions

$$T(0, \tau) = T_{\text{liq}}(0, \tau), \quad y_{i(j)}|_0 = y_{i(j)\text{inl}}, \quad \left.\frac{dy_{i(j)}}{dz}\right|_{0+z} = 0, \quad \left.\frac{dx_i}{dz}\right|_{0-z} = 0;$$

initial conditions

$$T(0) = T_{\text{liq}}(0) = T_{\text{liq.init}}; \quad x_i(0) = x_{i\text{init}}; \quad p_i(0) = x_i \exp(A_i - B_i/T_{\text{liq.init}}); \quad p_g = p_{\text{atm}}; \quad p_j(0) = 0; \quad j_{i(j)}(0) = 0.$$

The adequacy of the mathematical model is verified on an experimental setup which includes a chemical reactor, a capacitor, a water-jet ejector, a chromatograph, and devices for recording the temperature and absolute pressure.

Figure 1 presents the experimental data and calculated curves of change in the liquid-mixture temperature and partial pressures of the components as functions of the duration of a refining process of fats. By the
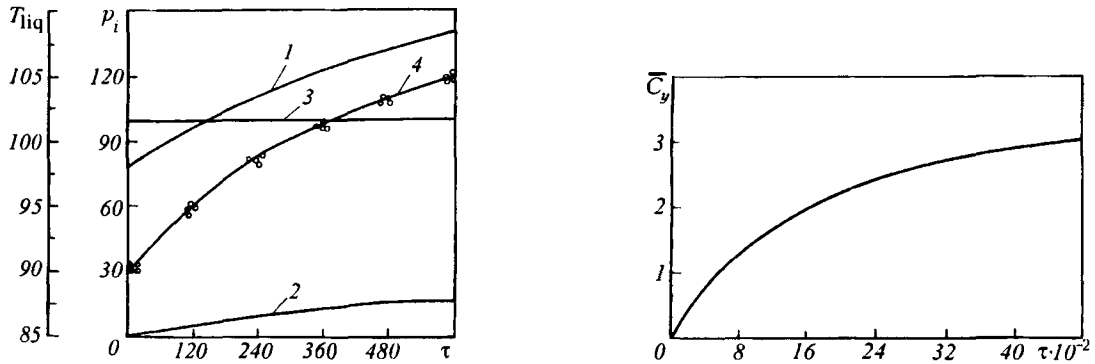
994

Fig. 1. Experimental data and calculated curves of change in the partial pressures of the components of a vapor-gas mixture: 1) steam; 2) vapor of sulfuric acid; 3) inert gas (air) in the reactor volume and in the liquid phase temperature (4) as functions of the duration of the fat-refining process. p_i, kPa; T_{liq}, K; τ, sec.

Fig. 2. Calculated kinetic curve of change in the mass concentration of tetrafluoroethylene in the free volume of the hermetic chamber in thermal decomposition of fluoroplastic-4. C_y, kg/m³.

technology of [15], one heats the fat with a live steam to a temperature of 363–368 K and introduces a concentrated sulfuric acid into it. Because of the condensate formed in heating, the acid is diluted to 30–40% and acts in this form on the impurities. As a result of the interaction of the liquids with different densities, the heat of dilution is liberated:

$$H_2SO_4 + nH_2O \rightarrow H_2SO_4 \cdot nH_2O + Q,$$
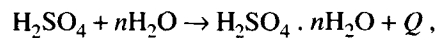
which favors the intense evaporation of the water and acid. The excess pressure in the reactor after 600 sec amounted to 140–150 kPa, and the temperature of the liquid mixture was increased up to 376–378 K.

The heat of dilution of the sulfuric acid can be approximately calculated from the Thomson equation [16]:

$$Q = 4.19 \left(\frac{17800n}{n + 1.7988} \right). \tag{32}$$

The mathematical model that includes Eqs. (14)-(16), (27), (29), and (32) is used in calculating and designing the equipment of gas purification [17] introduced at the "Nefis" Corporation, Kazan. The removal of generated vapor from the reaction zone by the suggested scheme eliminated the probability of its entry into the working zone.

The system of equations (6), (14)-(16), (22), and (31) is applied to the designing of a device [18] for the production of fluoroplastic cotton wool by means of laser decomposition of fluoroplastic-4. The decomposition of the polymer begins at a temperature of 695-698 K, while the entire liberating gas (tetrafluoroethylene) is a monomer [19]:

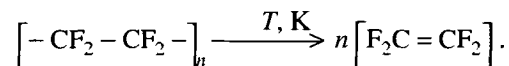$$\left[-CF_2 - CF_2 - \right]_n \xrightarrow{T, K} n \left[F_2C = CF_2 \right].$$

Figure 2 presents the calculated kinetic curve of change in the mass concentration of tetrafluoroethylene in the hermetic chamber. In the processing of one disk with a diameter of 80 mm and a thickness of 30 mm, up to 0.25 kg of a gaseous product capable of forming explosive and toxic products in oxidation is liberated for 1.3 h [20].

By the scheme suggested in [18], the decomposition process of the polymer occurs under vacuum conditions, while the trapping of the gas formed occurs in the ejector. In the interaction of tetrafluoroethylene with liquid bromine (which serves as a working medium), the gas is utilized by transforming it to 114V2-grade freon [20].

Moreover, based on a simultaneous solution of the internal and external problems, engineering solutions are developed [21-23] that provide environmental safety of technological processes for similar productions.

NOTATION

p, partial pressure of the component, Pa; p_w and p_m, pressure of the working liquid and surrounding medium, respectively, Pa; τ, time, sec; F, heat- and mass-transfer surface, m^2; R^*, universal gas constant, J/(kmol·K); E_{act}, activation energy, J/kmol; T and T_{liq}, temperature of the vapor-gas mixture and the liquid, K; j, mass flux, kg/(m^2·sec); $Q_{v.g}$ and Q_w, volumetric flow rates of the vapor-gas and working media, respectively, m^3/sec; V_f, free volume of the apparatus, m^3; μ, molecular weight of the component, kg/kmol; χ, order of the reaction in the reagent; c, mass heat capacity, J/(kg·K); c_μ, molar heat capacity, J/(kmol·K); $R_{v.g}$ and $R_{v.liq}$, reduced dimensions for the volumes with the vapor-gas and liquid media, respectively, m; k, constant of the homogeneous reaction rate, 1/(sec·$mol^{(\chi-1)}$); k', constant of the heterogeneous reaction rate, m/(sec·$mol^{(\chi-1)}$); k_0, preexponential factor; U_v, volumetric coefficient of injection; β, coefficient of mass transfer, m/sec; ρ, density, kg/m^3; y, mole-fraction of the component in the vapor, kmol/kmol; $\bar{y}$, mass fraction of the component in the vapor, kg/kg; $\bar{x}$, mass fraction of the component in the liquid, kg/kg; Q, heat of dilution, J; $q_{ch.r}$, specific heat of the chemical reaction, J/kg; α, coefficient of heat transfer, W/(m^2·K); S, interface, m^2; r, latent heat of vaporization, J/kg; A and B, empirical coefficients in the Antoine equation; φ, transfer potential; q, potential flux; w, rate, m/sec; γ, density of the potential source (sink); n, number of molecules, moles, and vapor components in the mixture and the polymerization depending on the context degree; m, number of gas components in the mixture; z, coordinate in the liquid–vapor system, m. Subscripts: liq, liquid; i, component of the vapor or the liquid; j, component of the gas; s, surface; f, free; v.g, vapor-gas mixture; sat, saturated; init, initial; cap, capacitor; μ, molar; inl, inlet; mix, mixture; g, gas; w, working; m, medium; v, volumetric; max, maximum; ch.r, chemical reaction; y, vapor-gas phase; act, activation; +z, vapor phase; −z, liquid phase; 0, phase interface.

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