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Temperature regime in a gas-drop mixture in the presence of an internal heat source

© N.M. Kortsenshtein

Joint Institute for High Temperatures, Russian Academy of Sciences,
125412 Moscow, Russia
e-mail: naumkor@yandex.ru

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The results of numerical simulation of the temperature regime of a mixture of gas and evaporating water droplets in a closed thermally insulated volume in the presence of an internal heat source are presented. The internal heat source is represented by a monomolecular exothermic reaction with temperature dependence of the rate constant according to the Arrhenius law. The influence of changing the initial values of gas and droplet temperatures, mass fraction and droplet radius on the temperature regime is investigated. The possibility of stabilization of the reacting gas temperature by evaporating droplets and limitations of the thermostating regime have been analyzed.

Keywords: droplets, evaporative cooling, exothermic reaction, Arrhenius law, numerical simulation.

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Introduction

The droplet evaporation process continues to attract attention, both scientific point of view and in connection with various applications [1]. In particular, evaporative cooling of solution droplets for obtaining nanoparticles [2,3], cooling of various surfaces with evaporating droplets [4–6] or ambient gas [7–9]. Relations were obtained for this case in [10,11], that relate the time of a decrease of gas temperature by a given amount to the initial values of gas temperature, mass concentration, and droplet radius. The control the temperature regime of an exothermic reaction by injecting highly dispersed water droplets into the reaction volume may be another application of the droplet evaporation process. This includes thermal processing of wood biomass [12]. The appearance of droplets in the reaction volume can have three effects: 1) the effect of the droplet surface on the course of the reaction; 2) the effect of the evaporating substance (vapor) on the course of the reaction; 3) the effect of the evaporation process on the temperature of the gas-droplet mixture and, as a result, on the course of the reaction. Attention is paid to the third effect in this paper. The greater the ratio of the heat of evaporation to the thermal effect of the reaction, the greater its significance. The temperature regime in a gas-droplet mixture is considered with the simultaneous course of two processes with respect to this effect — exothermic reaction and endothermic evaporation of droplets; the impact of the ratio of the characteristic times of these processes and the factors influencing this ratio is analyzed; the fundamental possibility of stabilizing the temperature of the reacting gas by evaporating droplets, as well as the limitation of the thermostatic control mode, is considered.

1. Problem statement and mathematical model

The gas with an internal heat source and droplets located in a closed thermally insulated volume are considered. The droplets are stationary, evenly distributed in volume and have the same size. The set values are the initial values of the temperature of the gas and droplets, the mass fraction and the radius of the droplets. It is assumed that the internal heat source is an exothermic monomolecular reaction in the gas phase of the form $B \rightarrow C$ with a temperature dependence of the reaction rate constant, according to the Arrhenius law, and the droplets and vapors formed do not affect the course of the reaction. Heat release during the reaction leads to an increase of the temperature of the gas phase, and interfacial heat exchange and subsequent evaporation of droplets leads to its decrease. The desired value is the time course of the temperature of the gas phase with simultaneous reaction and evaporation of droplets. The problem statement is illustrated in Fig. 1.

The solution to this problem is based on numerical modeling. The mathematical model used is similar to the model presented in Ref. [10] with additions that take into account the kinetics of the chemical reaction:

$$\frac{dg_B}{dt} = -k(T)g_B, \quad (1)$$

where g_B is the mass fraction of the component B , $k(T)$ is the reaction rate constant with temperature dependence according to the Arrhenius law:

$$k(T) = A \exp(-E/RT). \quad (2)$$

Here A , [s^{-1}] is the preexponential multiplier; E , [J/mol] is the activation energy, R , [J/(mol·K)] is the universal gas constant, T is the absolute temperature.

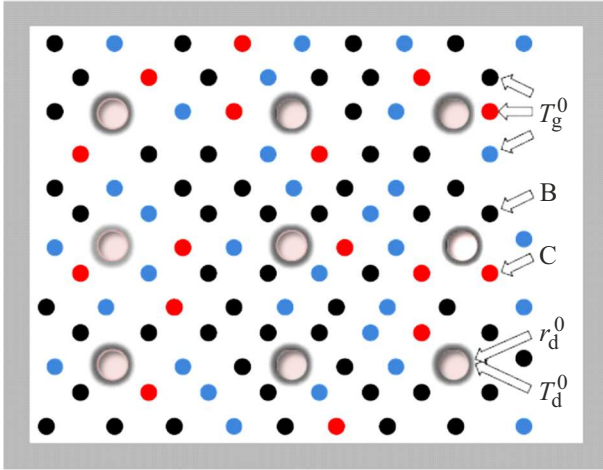


Figure 1. Problem statement scheme: a closed thermally insulated volume containing a mixture of gas, droplets and steam. Large circles — evaporating droplets. Small circles — molecules of the gas phase of grades B (black) and C (red), as well as vapor molecules (blue).

The equations of material balance in the processes of chemical transformation and evaporation of droplets relate changes in the mass fractions of the components of a mixture consisting of molecules of the grades B and C, steam (v) and droplets (d):

$$\frac{dg_C}{dt} = -\frac{dg_B}{dt}, \quad \frac{dg_v}{dt} = -\frac{dg_d}{dt}. \quad (3)$$

The material balance equation has the following form for droplets of the same size (monodisperse droplet size distribution)

$$\frac{dg_d}{dt} = \frac{3g_d}{r_d} \frac{dr_d}{dt}, \quad (4)$$

where g_i is the mass fraction of the corresponding component, r_d is the radius of the droplet.

The change of the mass fraction of all droplets is determined by the evaporation rate of one droplet as follows from equation (4). This statement is true for independently evaporating droplets when the average distance between the droplets ($n_d^{-1/3}$) is significantly larger than the droplet diameter, which limits the range of values g_d for which equation (4) applies. The mass fraction of droplets of the same size is determined by the expression

$$g_d = \frac{4}{3} \pi \rho_1 n_d r_d^3 / \rho_\Sigma. \quad (5)$$

Here n_d is the numerical droplet density, $[m^{-3}]$, droplet substance density, $[kg/m^3]$, ρ_Σ is the total density of a mixture of steam, gas and droplets, $[kg/m^3]$, which is a constant value in the considered case of a closed volume.

Taking into account (5) limitations on the applicability of equation (4) have the form

$$g_d^0 \leq \frac{\pi \rho_1}{6 \rho_\Sigma} N^{-3}, \quad (6)$$

$$n_d^0 \leq (2r_d^0 N)^{-3}, \quad (7)$$

where N is the ratio of the average distance between droplets to the diameter of the droplet. It can be seen that the limitation on the mass density does not depend on the droplet size. On the contrary, the limitations on the numerical density of droplets are determined precisely by their size. In particular, we obtain $n_d \leq 10^0 m^{-3}$ for droplets with a diameter of $100 \mu m$ at $N = 10$. The rate of evaporation of droplets, taking into account the Stefan flow, was calculated according to the expression [13] (see also [14]):

$$\frac{dr_d}{dt} = -\frac{\rho_{vg} D}{\rho_1 r_d} \left(\ln \frac{(1 - \alpha_1)}{(1 - \alpha_2)} \right). \quad (8)$$

Here

$$\alpha_1 = \frac{\rho_v}{\rho_{vg}}, \quad \alpha_2 = \frac{\rho_v^s}{\rho_{vg}}, \quad \rho_{vg} = \rho_v + \rho_g \frac{\mu_v}{\mu_g}, \quad (9)$$

where μ_i and ρ_i is the molar mass, $[kg/mol]$, and the density of the i -th component of the mixture, respectively, ρ_v^s is the density of saturated vapor, $[kg/m^3]$, D_v is the vapor molecule diffusion coefficient, $[m^2/s]$.

The energy equations for the components of the mixture have the form

$$c_g \frac{dT_g}{dt} = Q_{ex} \frac{dg_C}{dt} - Q_{gd}, \quad (10)$$

$$g_d c_d \frac{dT_d}{dt} = \frac{dg_d}{dt} (\Delta u_{vd}(T_d) + c_v(T_g - T_d)) + Q_{gd}. \quad (11)$$

Here T_g is the uniform gas temperature for the components of the gas phase,

$$c_g = g_{AC} c_A + g_{BC} c_B + g_v c_v \quad (12)$$

— heat capacity of the gas phase as a whole, $[J/(kg \cdot K)]$, c_i is the isochoric heat capacity of the i -th component, Q_{gd} , $[W/kg]$ is the amount of heat transferred per unit time in a unit mass of a mixture from gas to droplets, Q_{ex} , $[J/kg]$ is the thermal effect of a chemical reaction, Δu_{vd} is the change in internal energy in evaporation process $[J/kg]$.

Δu_{vd} was determined using a relationship with the evaporation heat L , $[J/kg]$, pressure p_v^s , $[PA]$, and density ρ_v^s of steam in the saturation state, $[kg/m^3]$:

$$\Delta u_{vd}(T) = L(T) - p_v^s(T) / \rho_v^s(T). \quad (13)$$

The following expression is used to determine the value of Q_{gd}

$$Q_{gd} = \left(\frac{3g_d}{\rho_1 r_d^2} \right) \frac{Nu}{2} \lambda_g (T_g - T_d), \quad (14)$$

where λ_g is the coefficient of thermal conductivity of the gas $[W/(m \cdot K)]$, Nu is the Nusselt number, $Nu = 2$ if the droplets are not blown by gas.

Accepted assumptions in the model:

1) The rapid equalization of vapor and gas temperatures in intermolecular collisions ($\tau_{vg} \sim 0.01 \mu s$) allows the use of one energy equation for the gas phase (at $t > \tau_{vg}$).

2) The temperature profile inside the evaporating droplets is uniform.

Let us find the ratio of the temperature equalization times in the droplet with a relative accuracy of less than 5% ($\tau_{0.05}^c$) and the droplet evaporation time (Δt_{vap}) to evaluate the correctness of this assumption. The Fourier criterion $Fo = 0.4$ corresponds to the desired value $\tau_{0.05}^c$ based on the data presented in Ref. [15]. Therefore

$$\tau_{0.05}^c = 0.4(r_d^0)^2/a_1, \quad (15)$$

where a_1 is the thermal conductivity of water, $[m^2/s]$. Let us use the following expression to estimate the evaporation time of a droplet [13]:

$$\Delta t_{vap} = \frac{\rho_1(r_d^0)^2}{2\rho_v^s D_v}. \quad (16)$$

As a result, we obtain

$$\frac{\tau_{0.05}^c}{\Delta t_{vap}} = 0.8 \frac{D_v \rho_v^s}{a_1 \rho_1}. \quad (17)$$

It can be seen that the desired ratio does not depend on the droplet radius, but is determined by the saturation line and the properties of vapor transfer and the droplet substance. This ratio has the values of 0.000635–0.0836 for water in the range from the triple point to the normal boiling point. It equals to 0.0124–0.0298 with the characteristic values of the temperature of the evaporating droplet in the heated gas of 323–343 K [10,11]. Consequently, the time of temperature equalization inside the droplet is much shorter than the time of evaporation of the droplet, and the assumption of a uniform temperature profile in the evaporating droplets is correct.

3) Droplet evaporation takes place in a quasi-stationary mode. We apply a similar approach to assess the correctness of this assumption. Let us determine the value of $\tau_{0.05}^D$, which ensures (at times exceeding $\tau_{0.05}^D$) the contribution of nonstationarity of less than 5% [13]:

$$\tau_{0.05}^D = 400r_d^2/\pi D_v. \quad (18)$$

The comparison of the values $\tau_{0.05}^D$ and Δt_{vap} yields

$$\frac{\tau_{0.05}^D}{\Delta t_{vap}} = \frac{800\rho_v^s}{\pi\rho_1}. \quad (19)$$

This ratio has the values of 0.0012–0.1523 for water in the range from the triple point to the normal boiling point. It equals to 0.0212–0.0517 with the characteristic values of the temperature of the evaporating droplet in the heated gas. 323–343 K [10,11]. Therefore, the assumption of a quasi-stationary evaporation regime is also correct.

4) Evaporation of each droplet occurs independently. This assumption and the limitations associated with it are discussed above.

5) Monodisperse droplet size distribution.

[16] compares the results of modeling of the evaporation of a collection of droplets in polydisperse and monodisperse approximations, as well as with experimental data from [17]. It is concluded that the discrepancy between the three groups of results is $\sim 10\%$. It should be noted that a similar conclusion was made in Ref. [18] when comparing the results of modeling heterogeneous vapor condensation on mono- and polydisperse centers.

The model was validated by using the data on gas cooling by evaporating droplets presented in Ref. [19]. As indicated in Ref. [19], the time required for gas cooling at $\Delta T = 20$ K in a gas droplet stream with an initial temperature of 296 K and a mass content of 3% of micron droplets (not specified more precisely in Ref. [19]), is $10 \mu s$. The same value, determined based on the numerical solution of the system of equations of the presented model for $r_d^0 = 1-2 \mu m$, gave a range of 3.5–14 μs . Considering the uncertainty of the value of r_d^0 , the result of using the model seems satisfactory.

2. Results and discussion

The model presented above was applied to analyze two cases: 1) the temperature regime in the reacting gas in the absence of droplets; 2) the temperature regime in the reacting gas in the presence of evaporating droplets. The proprietary program CONDKINET-1 was used for the numerical solution [20]. The thermophysical properties of reagents B and C were modeled by the properties of nitrogen. The necessary properties of the components (water, steam, nitrogen) were taken according to Ref. [21].

2.1. Temperature regime in reacting gas in absence of droplets

Characteristic values were selected for the parameters of the exothermic reaction $Q_{ex} = 74000$ J/kg, $E = 180$ kJ/mol, $A = 10^{13s-1}$ [12]. The initial gas temperature T_g^0 was the variable value. The calculation results are shown in Fig. 2.

It can be seen that, the reaction time greatly varies depending on the value of the initial temperature. At the same time, the temperature increase as a result of the reaction does not depend on the initial temperature and is determined by the balance ratio

$$T_g^{end} - T_g^0 = Q_{ex}/c_g, \quad (20)$$

where T_g^{end} is the final temperature of the gas.

The equations (1) and (10) can be reduced to the same form with the same zero initial conditions in the absence of droplets

$$\frac{d\xi}{dt} = k(T_g)(1 - \xi), \quad (21)$$

$$\frac{d\theta}{dt} = k(T_g)(1 - \xi). \quad (22)$$

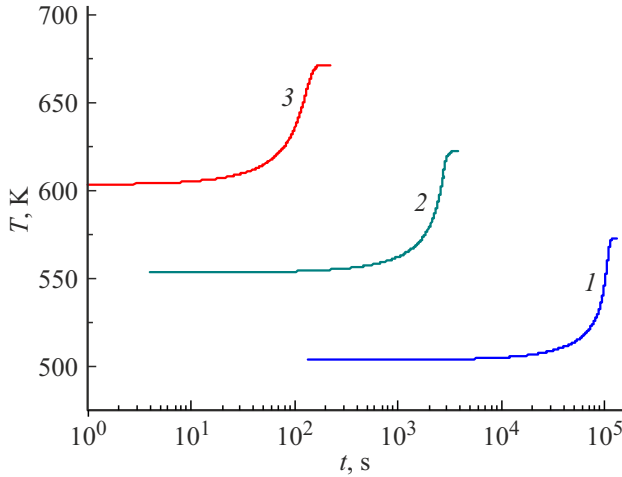


Figure 2. Time dependence of gas temperature at different values T_g^0 : 1 — 503.15 K, $E/(RT_g^0) = 43.03$; 2 — 553.15 K, $E/(RT_g^0) = 39.14$; 3 — 603.15 K, $E/(RT_g^0) = 35.90$.

Here ξ is the degree of transformation, defined as

$$\xi = 1 - \frac{g_B}{g_B^0} \quad 0 \leq \xi \leq 1, \quad (23)$$

g_B^0 — mass fraction of component B at the initial time, θ — dimensionless temperature:

$$\theta = \frac{T_g - T_g^0}{T_g^{\text{end}} - T_g^0} = \frac{c_g(T_g - T_g^0)}{Q_{\text{ex}}} \quad 0 \leq \theta \leq 1. \quad (24)$$

The solution of equation (22) has the following form at the initial stage of the reaction, with a small degree of transformation ($\xi \ll 1$)

$$\theta = k(T_g^0)t. \quad (25)$$

The obtained solution makes it possible to determine the heating time of the reacting gas to a certain level at the initial stage of the reaction, depending on the initial temperature of the gas:

$$t = \frac{\theta}{k(T_g^0)} \quad (26)$$

or

$$\tau = \theta. \quad (27)$$

Here τ is the dimensionless time

$$\tau = tk(T_g^0). \quad (28)$$

A comparison of the heating time obtained from the numerical solution of equation (22) and the approximate analytical solution (27) is shown in Fig. 3. It can be seen that the approximate analytical solution (27) gives an upper estimate for the heating time. The error of determining the warm-up time using the expression (27) does not exceed 10% at $\theta \leq 0.05$.

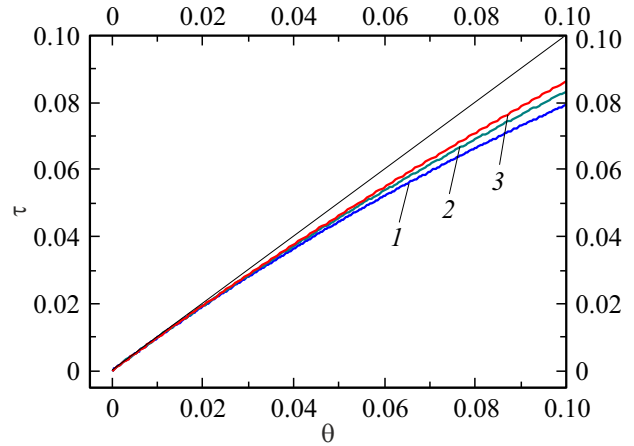


Figure 3. Dependence of the time of gas heating to the final temperature in dimensionless coordinates for various values T_g^0 . Notation of curves according to Fig. 2. The straight line corresponds to the dependency $\tau = \theta$.

2.2. Temperature regime in a reacting gas in the presence of evaporating droplets

There is a competition between two processes in this case: exothermic reaction in the gas phase and endothermic evaporation of droplets. It can be assumed that one of three scenarios is realized depending on the ratio of the characteristic evaporation times Δt_{vap} and the reaction Δt_r : the reaction has time to end and heat the gas before evaporation begins under the condition $\Delta t_{\text{vap}} \gg \Delta t_r$ and then the evaporating droplets cool it; the droplets have time to evaporate at $\Delta t_{\text{vap}} \ll \Delta t_r$ and cool the practically non-reacting gas, in which the reaction then begins; both processes occur simultaneously and influence each other at $\Delta t_{\text{vap}} \sim \Delta t_r$.

The temperature dependence of $\Delta t_{\text{vap}}/\Delta t_r$ is shown in Fig. 4. The characteristic time of the monomolecular reaction was determined as

$$\Delta t_r = k(T_g^0)^{-1}, \quad (29)$$

and the time of the evaporation process was found from the numerical solution of the system of equations of the model presented above without taking into account the thermal effect of the reaction. Fig. 4 shows that it is possible to implement all three scenarios with varying initial droplet radius at sufficiently high temperatures corresponding to the condition $(E/(RT_g^0))$. On the contrary, only the scenario of advanced evaporation of droplets is realized at sufficiently low temperatures, when $(E/(RT_g^0) > 32)$. The observed behavior of the value $\Delta t_{\text{vap}}/\Delta t_r$ is associated with two circumstances: the quadratic dependence of the evaporation time of droplets on their radius (see [13]) and the exponential dependence of the reaction rate constant (and the characteristic reaction time) on temperature (see expressions (2) and (29)). The first circumstance is crucial

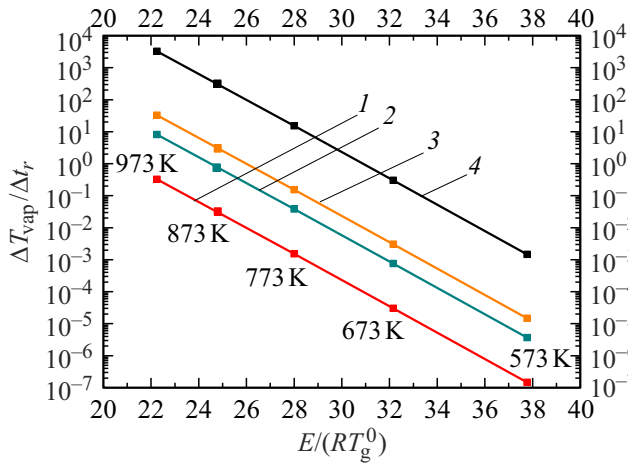


Figure 4. The ratio of the characteristic evaporation and reaction times depending on the initial values of the gas temperature and the droplet radius: 1 — 1, 2 — 5, 3 — 10, 4 — 100 μm . Initial droplet temperature $T_d^0 = 293.15 \text{ K}$.

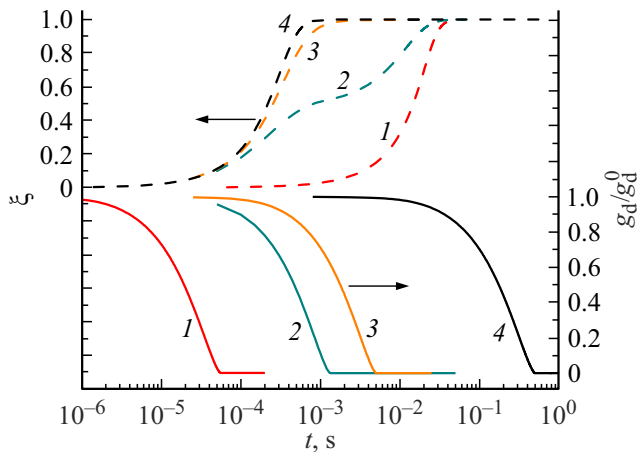


Figure 5. Dependence of the degree of completion of the reaction (dashed curves) and the droplet evaporation process (solid curves) depending on the time at different initial values of the droplet radius: 1 — 1, 2 — 5, 3 — 10, 4 — 100 μm . $T_d^0 = 293.15 \text{ K}$, $g_d^0 = 5\%$.

at high temperatures ($E/(RT_g^0) < 24$), the second circumstance is crucial at low temperatures ($E/(RT_g^0) > 32$).

The data of numerical integration of the model's system of equations presented in Fig. 5–8 confirm the qualitative considerations presented above. As follows from Fig. 5–6, the variation of the initial radius of the droplets (respectively, their evaporation time) at a constant initial gas temperature allowed tracing the transition from one scenario to another.

It can be seen that the curves 1 in Fig. 5, 6 correspond to advanced evaporation of droplets, and the curves 4 correspond to advanced reaction. The curves 2 and 3 correspond to simultaneous processes. The ratio $\Delta t_{\text{vap}} = \Delta t_r/3$ was sufficient in this case for the predominance of the droplet evaporation process. It should be noted (Fig. 6) that

for all variants with the same value of the initial droplet concentration, the final gas temperature after evaporation of the droplets is the same, as it should be according to the thermal balance condition.

The data shown in Fig. 7, 8, demonstrate the influence of the initial values of the mass fraction of droplets and the temperature of the gas on the time course of the gas temperature and the degree of completion of the reaction. It can be seen (Fig. 7) that an increase of the initial concentration of droplets increases the reaction time due to more intensive cooling of the gas by evaporating droplets and the reaction at a lower temperature. Fig. 8 shows that a qualitative similarity of the time dependences of the gas temperature is achieved with lower values of the initial droplet concentration with a decrease of the initial

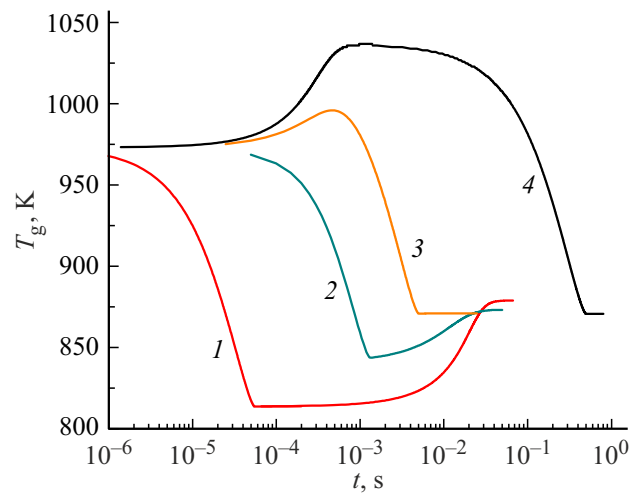


Figure 6. Dependence of gas temperature on time at different initial values of the droplet radius: 1 — 1, 2 — 5, 3 — 10, 4 — 100 μm . $T_d^0 = 293.15 \text{ K}$, $T_g^0 = 973.15 \text{ K}$, $g_d^0 = 5\%$.

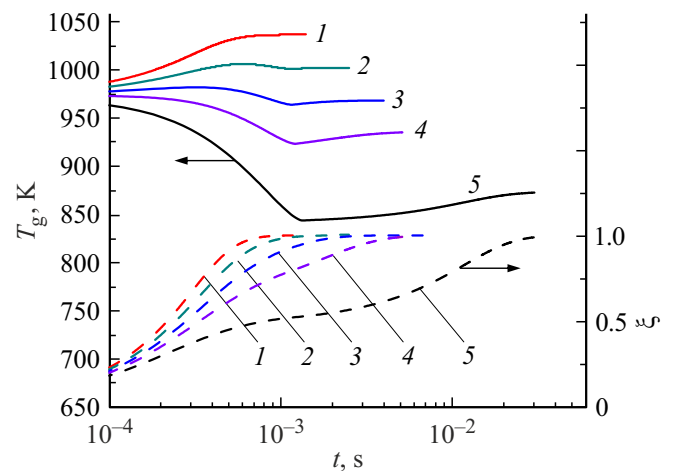


Figure 7. Dependence of the gas temperature and the degree of completion of the reaction during droplet evaporation on the time and initial mass fraction of droplets: 1 — 0, 2 — 1, 3 — 2, 4 — 3, 5 — 5%. $T_g^0 = 973.15 \text{ K}$, $r_d^0 = 5 \mu\text{m}$.

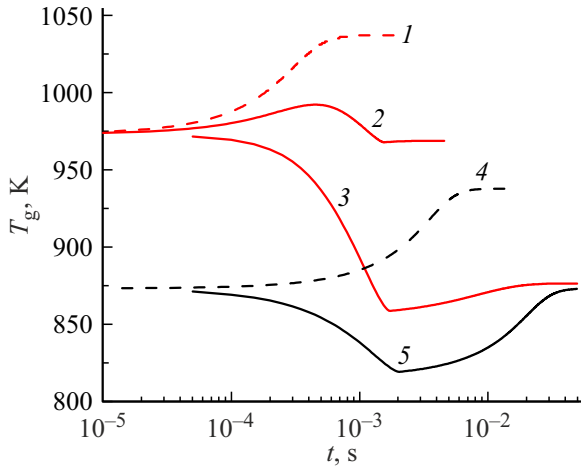


Figure 8. The dependence of the gas temperature on time for different values of the mass fraction of droplets and the initial temperature of the gas: 1, 4 — no droplets; 2, 5 — 2%; 3 — 5%. 1–3 — 973.15 K; 4, 5 — 873.15 K.

gas temperature. In particular, the gas cooling mode with evaporating droplets followed by the reaction is achieved at $T_g^0 = 973.15$ K if the initial mass fraction of droplets is 5% (Fig. 8, curve 3). The same mode is realized at $T_g^0 = 873.15$ K if the initial mass fraction of droplets is 2% (Fig. 8, curve 5).

2.3. About the stabilization of the temperature of the reacting gas by evaporating droplets

As noted above, only the scenario of advanced evaporation of droplets is realized at sufficiently low temperatures, when $(E/(RT_g^0)) > 32$. Therefore, the droplet evaporation process can be used to maintain the temperature of the reacting gas within specified limits in this case, since the time of cooling of the gas by evaporating droplets is significantly less than the time of heating of the gas due to the reaction. Let us consider the fundamental possibility of a cyclic process according to the scheme: gas heating without droplets — droplet injection followed by gas cooling by evaporating droplets. Let us call this the temperature control mode. Let the permissible value of the gas temperature be $T_{g \max}$. Wherein $T_{g \max} - T_g^0 \ll T_g^0$. The corresponding value is $\theta_{\max} \ll 1$ in this case according to (24). The heating time of the gas without droplets to the set limit value is according to (26):

$$\Delta t_{\max} = \theta_{\max} / k(T_g^0). \quad (30)$$

At the same time, Δt_{\max} determines the lifetime of the gas in a given temperature range of θ_{\max} . It should be noted that this time is shorter the higher the gas temperature to be stabilized.

The mass concentration of droplets for evaporative cooling of the gas to the initial temperature is determined from

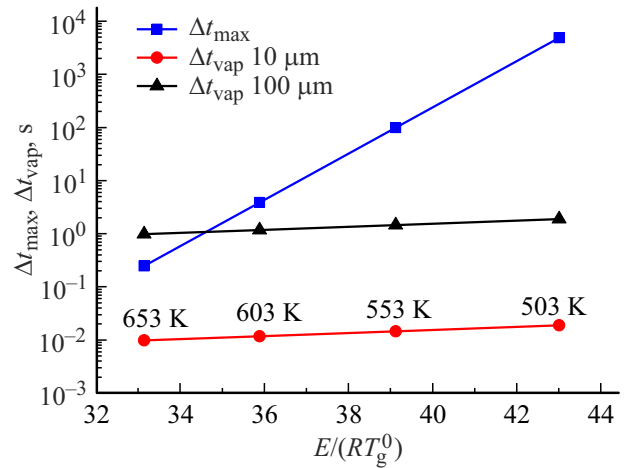


Figure 9. The times of gas heating and evaporative cooling, depending on the initial values of the gas temperature and the droplet radius.

the balance ratio

$$g_{d \max} = \frac{c_g(T_{g \max} - T_g^0)}{\Delta u_{vd}} = \theta_{\max} \frac{Q_{ex}}{\Delta u_{vd}}, \quad (31)$$

where Δu_{vd} is the change of internal energy during evaporation, [J/kg] (see expression (13)). We obtain $g_{d \max} \approx 3 \cdot 10^{-4}$ using $\theta_{\max} = 0.01$ for estimates with the selected value Q_{ex} . Evaporation of a certain number of droplets according to (31) will ensure that the gas temperature returns to its original value T_g^0 — the cycle will close. Obviously, the condition $\Delta t_{vap} \ll \Delta t_{\max}$ should be met to implement the temperature control mode. The required droplet injection frequency will be $(\Delta t_{\max})^{-1}$ when this condition is met. The results of the determination of Δt_{vap} based on numerical modeling and calculations of Δt_{\max} , according to (26), are shown in Fig 9.

The discussed method of temperature stabilization of the reacting gas is possible in principle as can be seen from Fig. 9. There is a temperature range where the required ratio of heating and evaporative cooling times is met. Two circumstances determine the upper temperature limit of this region. Firstly, an increase of the initial temperature reduces the lifetime of the gas in a given temperature range and, consequently, increases the required frequency of droplet injection; secondly, an increase in the initial temperature of the gas brings the heating and evaporation times closer. An increase of the initial radius of the droplets shifts the upper temperature boundary to a region of lower temperatures. There is also a lower temperature limit, which is associated with a slowdown of the reaction rate as the temperature decreases.

Conclusion

The thermal mechanism of interaction between an exothermic reaction and evaporating droplets is considered

based on numerical modeling. It was found that changes of the initial temperatures of the gas and droplets, the mass fraction and the radius of the droplets affect the temperature regime due to changes in the characteristic reaction times and evaporation of droplets. In particular:

1) the variation of the initial radius of droplets and their mass fraction (respectively, their evaporation time) at a constant initial gas temperature allows switching from the mode of advanced reaction to the mode of advanced evaporation of droplets with a corresponding change of temperature dependence on time in the region of sufficiently high initial gas temperatures corresponding to the condition $(E/(RT_g^0) < 24)$;

2) an increase of the initial mass fraction of droplets increases the reaction time due to more intensive cooling of the gas by evaporating droplets and the reaction at a lower temperature;

3) a qualitative similarity of the time dependences of the gas temperature is achieved with a decrease of the initial temperature of the gas with lower values of the initial mass fraction of droplets;

4) only the mode of advanced evaporation of droplets is implemented in the region of sufficiently low initial gas temperatures, when $(E/(RT_g^0) > 32)$, which makes it possible to control the temperature of the reacting gas within the specified limits by evaporating droplets;

5) The temperature control mode has a higher and lower temperature limitation.

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Conflict of interest

The author declares that he has no conflict of interest.

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