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DETERMINATION OF THE CHARACTERISTICS OF NONSTEADY HEAT EXCHANGE IN ONE DISPERSE REACTIVE SYSTEM

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The laws of nonsteady heat exchange in a disperse reactive system are investigated within the framework of a new model of heat exchange in a disperse medium not using the concept of a coefficient of heat exchange.

The problem of energy transfer in stationary, disperse, nonreacting and reacting media has been investigated in [1-7]. An analysis of the heat-transfer mechanism is given in these reports and the basic equations are obtained. In [2], in particular, a review is made of the methods for mathematical modeling of the heat exchange in disperse media and it is pointed out that the simplest means of mathematical modeling is the use of the ordinary heat-conduction equations and the volume heat-transfer coefficients. A sufficiently simple two-temperature model for the heat exchange in disperse media is obtained as a result. A more complicated mathematical model of the transfer processes in a reacting medium, allowing for the multiphase and multitemperature nature of the medium, is suggested in [4]. In this model, however, they use the concept of the heat-transfer coefficient, which is a function of time not known in advance, and additional assumptions are introduced in connection with the use of so-called accomodation equations.

The problem of the nonsteady heat exchange in a disperse system is analyzed below within the framework of the mathematical model given in [8].

Suppose there is a vessel filled with a liquid or gaseous substance whose temperature is known and equal to T_n. A constant temperature T_n is maintained at the vessel walls during the entire process. Reactive spherical particles of a solid substance, equal in mass and having the same initial temperature T_{in}, enter the vessel at some moment. We assume that the particles will be in a suspended state at equal distances from each other and that chemical reactions whose rates are determined by the Arrhenius law [9] can be observed at the particle—gas (liquid) interface. We presume that the depletion of the material of the particles during their ignition is small, so that their depletion and the depletion of the gas is ignored. Moreover, we assume that the following assumptions are valid:

1) the number concentration of particles per unit volume of disperse medium is known and equal to n;

2) the process of heat transfer as a result of molecular heat conduction is one-dimensional;

3) the thermophysical coefficients of the particles and gas are constant;

4) the temperature inside any particle does not vary from point to point, since the radius of a particle is small.

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From symmetry considerations it follows that an equal volume of the carrier medium, having the shape of a cube, surrounds each particle. The size b of a cube edge can be determined from the equation

$$b = (n)^{-1/3}.$$

Because of symmetry the derivative of the temperature along a normal at its faces is equal to zero.

Thus, the problem of the "internal" heat exchange in the given disperse system comes down to the problem of the heat exchange between a particle and the carrier medium within the indicated cube. Owing to the nonlinearity of the equations entering into it and the three-dimensionality of the region of its determination, however, its solution is connected with great computational difficulties.

The solution of this problem can be simplified considerably if one employs the so-called principle of heat-flux stability [10], analogous to the St. Venant principle which is well known in the theory of elasticity. The principle of heat-flux stability specifies that any local disturbance of the temperature field is localized and does not extend to remote parts of the field if the heat flux through the surface of the body remains constant. An estimate of the error of the principle of heat-flux stability is given for some heat-conduction problems in [10], where the cooling curves for a cube and an equivalent sphere were compared, in particular. A sphere is called equivalent which is composed of the same material as the cube with the volume of the sphere being equal to the volume of the cube and the same boundary conditions being satisfied at the surface of the sphere as at the faces of the cube. It is asserted [10] that the error of the stability principle is small and in the central part of a cube the isotherms have an almost spherical shape.

When the cube is replaced by an equivalent sphere the solution of the problem of the heat exchange between the particle and the carrier medium is simplified considerably, since in this case the heat-conduction equation becomes one-dimensional. To formulate the boundary conditions one introduces the concept of the radius r_e of the cube, by which one understands the radius of the sphere equivalent to the cube.

For a cube inside which a typical particle is located the equivalent radius is

$$r_e = \left(\frac{3b^3}{4\pi}\right)^{1/3} = \left(\frac{3}{4\pi n}\right)^{1/3}.$$

By virtue of the assumptions made, the solution of the problem of self-ignition of the disperse system under consideration comes down to the solution of the equation of heat conduction in a disperse medium and the equations of heat conduction within the equivalent spheres:

$$\left\{c_1\rho_1\left[1-\left(\frac{r_e}{r_2}\right)^{-3}\right]+c_2\rho_2\left(\frac{r_e}{r_2}\right)^{-3}\right\}\frac{\partial T}{\partial t}=\frac{1}{y^k}\frac{\partial}{\partial y}\left[y^k\lambda\frac{\partial T}{\partial y}\right]-4\pi r_2^2\lambda_1 n\left.\frac{\partial T_1}{\partial r}\right|_{r=r_2},\quad 0<y<l, \tag{1}$$

$$c_1\rho_1\frac{\partial T_1}{\partial r}-\frac{\lambda_1}{r_2^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial T_1}{\partial r}\right)=0,\quad r_2<r<r_e. \tag{2}$$

Equation (2) is the heat-conduction equation for a typical equivalent sphere.

The boundary conditions and initial conditions for (1), (2) have the form

$$\left.\frac{\partial T}{\partial y}\right|_{y=0}=0,\quad T|_{y=l}=T_n.\ T_1|_{r=r_e}=T(y,\ t),\quad T_1|_{r=r_2}=T_0, \tag{3}$$

$$\frac{1}{3}\rho_2c_2\frac{dT_0}{dt}=\lambda_1\left.\frac{dT_1}{dr}\right|_{r=r_2}+qK_0\exp\left(-\frac{E}{RT_0}\right), \tag{4}$$

$$t=0,\quad T=T_1=T_0=T_n. \tag{5}$$

Equation (4) represents the equation of thermal energy balance in a typical particle, while the y coordinate in the third condition of (3) plays the role of a parameter.

According to [8], to satisfy the third of the conditions (3) it is enough that one of the following inequalities be satisfied:

$$r_e^2/\varkappa \ll l^2/\varkappa_s,\quad \varkappa=\lambda/\rho c_p, \tag{6}$$

$$r_e^2/\varkappa \gg l^2/\varkappa_s. \tag{7}$$

When (6) is satisfied the "internal" heat exchange is almost complete, as a result of which the temperatures of the gas within the equivalent sphere and of the particle differ little from each other. Conversely, when the inequality (7) is satisfied the thermal wave is not able to reach the boundary of the equivalent sphere and the initial temperature at its boundary remains. Intermediate regions of variation of the characteristic times of thermal relaxation are realized most often in practice, so that a final judgement about the adequacy of the model and its nature can be made in each concrete case on the basis of a comparison of theoretical and experimental data.

Equations (1), (2), and (4) were obtained on the basis of equations suggested in [8]. The quantity λ was calculated from the equations of [11].

For generality of the solution we rewrite the boundary problem (1)-(5) in dimensionless variables:

$$\frac{\partial}{\partial \eta}\left(\eta^k \frac{\partial \theta}{\partial \eta}\right) = \eta^k \left(A \frac{\partial \theta}{\partial \tau} + B \frac{\partial \theta_1}{\partial x}\bigg|_{x=1}\right), \quad 0 < \eta < \pi_l, \tag{8}$$

$$\frac{\partial \theta_1}{\partial \tau} = \frac{1}{\delta_2 x^2} \frac{\partial}{\partial x}\left(x^2 \frac{\partial \theta_1}{\partial x}\right), \quad 1 < x < x_e. \tag{9}$$

The initial and boundary conditions for the system of equations (8), (9) are written in the following form:

$$\text{at} \quad \tau = 0: \quad \theta = \theta_n = \theta_1 = \bar{\theta}_0 = 0, \tag{10}$$

$$\frac{\partial \theta}{\partial \eta}\bigg|_{\eta=0} = 0, \quad \theta|_{\eta=\pi_l} = \theta_n, \quad \theta_1|_{x=x_e} = \theta(\eta, \tau), \quad \theta_1|_{x=1} = \bar{\theta}_0, \tag{11}$$

$$\frac{d\bar{\theta}_0}{d\tau} = \frac{3\alpha}{\delta_2} \frac{\partial \theta_1}{\partial x}\bigg|_{x=1} + 3\exp\frac{\bar{\theta}_0}{1+\beta\bar{\theta}_0}, \tag{12}$$

where

$$\theta_1 = \frac{(T_1 - T_*)E}{RT_*^2}; \quad \theta = \frac{(T - T_*)E}{RT_*^2}; \quad \bar{\theta}_0 = \frac{(T_0 - T_*)E}{RT_*^2};$$

$$\theta_w = \frac{(T_w - T_*)E}{RT_*^2}, \quad k = 0, 1, 2;$$

$$\eta = \frac{y}{r_2}; \quad \tau = \frac{t}{t_*}; \quad t_* = \frac{c_2\rho_2}{qk_0\rho_1}\exp\frac{E}{RT_*};$$

$$A = \frac{\delta_1[1 - (x_e)^{-3} + \alpha^{-1}(x_e)^{-3}]}{\bar{\lambda}};$$

$$B = \frac{3\delta_1(x_e)^{-3}}{\delta_2\lambda}; \quad x_e = \frac{r_e}{r_2}; \quad \alpha = \frac{c_1\rho_1}{c_2\rho_2}; \quad \pi_l = \frac{l_0}{r_2}, \quad \delta_2 = \frac{r_2^2 c_1\rho_1}{\lambda_1 t_*};$$

$$\bar{\lambda} = \frac{\pi_\lambda - (\pi_\lambda - 1)[1 - (x_e)^{-2/3}](x_e)^{-1/3}}{\pi_\lambda - (x_e)^{-1/3}(\pi_\lambda - 1)}; \quad \pi_\lambda = \frac{\lambda_2}{\lambda_1}, \quad \delta_1 = \frac{l^2 c_1\rho_1}{\lambda_1 t_*},$$

$$\beta = \frac{RT_*}{E}.$$

The problem is solved using the iteration–interpolation method [12]. Since the conditions for ignition are the most favorable at the center of the vessel, we henceforth consider the heat exchange between any particle at the center of the vessel and the surrounding medium.

In accordance with [12], we assign the function θ in the form

$$\theta = a\eta^2 + b.$$

The quantities a and b are determined from the conditions (11): $b = \theta_w$ and $a = \theta_n - \theta_w$, where θ_w is the temperature of the disperse medium at $\eta = 0$. Then

$$\theta = \theta_n \eta^2 + \theta_w (1 - \eta^2). \tag{13}$$

The temperature θ_1 of the carrier medium depends on the coordinate η as on a parameter. We assign this dependence in the form

$$\theta_1 = \theta_n \eta^2 + \theta_0 (1 - \eta^2), \tag{14}$$

where θ_0 is the dimensionless temperature of the carrier medium at the center of the vessel. Substituting (13) and (14) into (8) and integrating once, we obtain

$$\frac{\partial \theta}{\partial \eta} = A \dot{\theta}_w \left(\frac{\eta}{k+1} - \frac{\eta^3}{k+3} \right) + B \left. \frac{\partial \theta_1}{\partial x} \right|_{x=1} \left(\frac{\eta}{k+1} - \frac{\eta^3}{k+3} \right) + \frac{c_1}{\eta^k}. \tag{15}$$

From the first condition of (11) it follows that $c_1 = 0$. Integrating (15), we obtain

$$\theta = A \dot{\theta}_w \left[\frac{\eta^2}{2(k+1)} - \frac{\eta^4}{4(k+3)} \right] + B \left. \frac{\partial \theta_1}{\partial x} \right|_{x=1} \left[\frac{\eta^2}{2(k+1)} - \frac{\eta^4}{4(k+3)} \right] + c_2. \tag{16}$$

From the requirement that expression (16) for θ satisfy the boundary conditions (11) and (12) we obtain, after obvious transformations, the ordinary differential equation

$$\left[A \dot{\theta}_w + B \left(\dot{\theta}_0 - 3 \exp \frac{\theta_0}{1 + \beta \theta_0} \right) \frac{\delta_2}{3\alpha} \right] \frac{k+5}{4(k+1)(k+3)} = \theta_n - \theta_w. \tag{17}$$

We apply the method of [12] to Eq. (9). We assign the dimensionless temperature θ_1 in the form

$$\theta_1 = A_1 x + B_1. \tag{18}$$

From the fact that (18) satisfies the boundary conditions (11), we find

$$A_1 = \frac{\theta_0 - \theta_w}{1 - x_e}, \quad B_1 = \frac{\theta_w - x_e \theta_0}{1 - x_e}. \tag{19}$$

Substituting (18) into (9), we obtain

$$\frac{\partial}{\partial x} \left(x^2 \frac{\partial \theta_1}{\partial x} \right) = \delta_2 x^2 \left(\frac{\dot{\theta}_0 - \dot{\theta}_w}{1 - x_e} x + \frac{\dot{\theta}_w - \dot{\theta}_0 x_e}{1 - x_e} \right). \tag{20}$$

Integrating (20) with respect to x, we obtain

$$\frac{\partial \theta_1}{\partial x} = \delta_2 \left[\frac{(\dot{\theta}_0 - \dot{\theta}_w)}{1 - x_e} \frac{x^2}{4} + \frac{(\dot{\theta}_w - \dot{\theta}_0 x_e)}{1 - x_e} \frac{x}{3} \right] + \frac{d_1}{x^2}. \tag{21}$$

Using the condition (10), we find

$$d_1 = \left(\dot{\theta}_0 - 3 \exp \frac{\theta_0}{1 + \beta \theta_0} \right) \frac{\delta_2}{3\alpha} - \delta_2 \left[\frac{\dot{\theta}_0 - \dot{\theta}_w}{4(1 - x_e)} + \frac{\dot{\theta}_w - \dot{\theta}_0 x_e}{3(1 - x_e)} \right].$$

Integrating (21) again, we obtain

$$\theta_1 = \delta_2 \left[\frac{\dot{\theta}_0 - \dot{\theta}_w}{1 - x_e} \frac{x^3}{12} + \frac{(\dot{\theta}_w - \dot{\theta}_0 x_e)}{1 - x_e} \frac{x^2}{6} \right] - \frac{d_1}{x} + d_2, \tag{22}$$

where

$$d_2 = \theta_0 + d_1 - \delta_2 \left[\frac{\dot{\theta}_0 - \dot{\theta}_w}{12(1 - x_e)} + \frac{(\dot{\theta}_w - \dot{\theta}_0 x_e)}{6(1 - x_e)} \right].$$

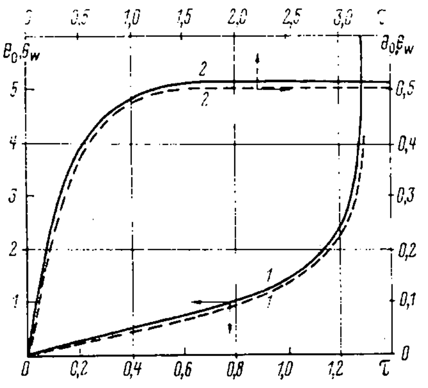

Fig. 1

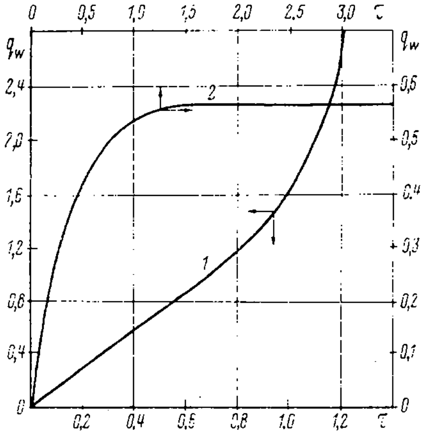

Fig. 2

Fig. 1. Time dependences of dimensionless particle temperature θ_0 (solid curves 1 and 2) and temperature θ_w of disperse medium (dashed curves 1 and 2) on time at self-ignition (curves 1; $D = 1.7$; $\delta_1 = 0.062$; $\delta_2 = 0.62\cdot10^{-5}$; $k = 2$; $\alpha = 0.67\cdot10^{-3}$; $x_e = 3$; $\pi_\lambda = 3472$) and at emergence into steady-state mode (curves 2; $D = 3.2$; $\delta_1 = 0.062$; $\delta_2 = 0.62\cdot10^{-5}$; $k = 2$; $\alpha = 0.67\cdot10^{-3}$; $x_e = 4$; $\pi_\lambda = 3742$).

Fig. 2. Time dependences of dimensionless heat flux q_w during self-ignition (1) and emergence into steady-state mode (2). Values of dimensionless parameters same as for Fig. 1.

From the fact that Eq. (22) satisfies the boundary condition $\theta_1|_{x=x_e} = \theta_w$ we find the second ordinary differential equation:

$$\dot{\theta}_0\left[-\frac{5}{12}+\frac{1}{3\alpha}+\frac{1}{x_e}\left(\frac{1}{4}-\frac{1}{3}\alpha\right)+\frac{x_e}{12}+\frac{x_e^2}{12}\right]+\dot{\theta}_w\left[\frac{1}{12x_e}-\frac{1}{12}-\frac{x_e}{12}+\frac{x_e^2}{12}\right]=\frac{\theta_w-\theta_0}{\delta_2}+\frac{x_e-1}{\alpha x_e}\exp\frac{\theta_0}{1+\beta\theta_0}. \tag{23}$$

As a result, we obtain two ordinary differential equations, (17) and (23), for the determination of the functions θ_0 and θ_w. The initial conditions for (17) and (23) have the form

$$\theta_w(0)=0,\quad \theta_0(0)=0. \tag{24}$$

From the physical statement of the problem, it follows that the system under consideration can self-ignite at certain values of the thermokinetic parameters and certain heat-exchange conditions. It is therefore interesting to determine the critical conditions for self-ignition.

The critical conditions for ignition are determined by the limiting values of the system parameters at which a steady-state solution of Eqs. (17) and (23) is absent. Taking $d\theta_w/d\tau \equiv 0$ and $d\theta_0/d\tau \equiv 0$ in (17) and (23), we obtain the steady-state equations:

$$-\frac{B\delta_2}{\alpha}\exp\frac{\theta_0}{1+\beta\theta_0}=\frac{(\theta_n-\theta_w)\,4\,(k+1)(k+3)}{k+5}, \tag{25}$$

$$-\frac{(x_e-1)}{\alpha x_e}\exp\frac{\theta_0}{1+\beta\theta_0}=\frac{\theta_w-\theta_0}{\delta_2}. \tag{26}$$

From (25) and (26) we find one equation for the determination of the dimensionless particle temperature θ_0:

$$\frac{1}{\delta_2}\left[\frac{m\theta_n(x_e-1)}{Bx_e+m(x_e-1)}-\frac{\theta_0(x_e-1)\,m}{Bx_e+m(x_e-1)}\right]=-\frac{(x_e-1)}{\alpha x_e}\exp\frac{\theta_0}{1+\beta\theta_0}, \tag{27}$$

where $m = 4(k + 1)(k + 3)/(k + 5)$.

Since the temperature maintained at the vessel surface is adopted as the temperature scale T_*, we have $\theta_n = 0$. Then from (27) at $\beta = 0$,

$$\frac{m\alpha x_e}{\delta_2\,[Bx_e + m(x_e - 1)]} = \frac{\exp\theta_0}{\theta_0} = D. \tag{28}$$

The quantity D has a minimum with respect to θ_0 at the point $\theta_0 = 1$. Values of $D > e$ correspond to real values of the steady temperature θ_0 of a typical particle, while at $D < e$ Eq. (27) has no real solution. Consequently, the parameter D can be taken as the value determining the limiting values of the system parameters at which self-ignition is absent. At $D < e$ the system ignites while at $D \geq e$ it does not ignite.

The result is confirmed by numerical integration of (17) and (23).

In Fig. 1 we present values of the particle temperature θ_0 (solid curves 1 and 2) and the temperature θ_W of the disperse medium (dashed curves 1 and 2) for values of the parameter $D < e$ and $D > e$. As follows from the figure, the temperature of a typical particle which is at the center of the vessel grows smoothly from the time $\tau = 0$ to $\tau = 1$ while curve 1 goes upward steeply at $\tau > 1$. The time $\tau = \tau_*$ at which the temperature θ_0 emerges onto the vertical asymptote is taken as the self-ignition time. By the time $\tau = \tau_*$ the temperature θ_W of the disperse medium also emerges onto the vertical asymptote, but the values of θ_W are less than those of θ_0 as this happens.

This result agrees in a qualitative respect with the data of [7].

Curves 2 in Fig. 1 are typical graphs of the temperatures θ_0 and θ_W of the particle and the disperse medium at $D > e$. In the given case temperatures θ_0 and θ_W which are close in value are established with time, and they do not vary with time and differ little from zero. The thermal energy released by the reacting particles is removed from the vessel by heat conduction. In this case a steady mode of occurrence of the process takes place and ignition does not happen.

Curves of the heat flux q_W to the vessel wall for nonsteady heat exchange and for the case of nonignition are presented in Fig. 2. As is seen, by the moment of ignition the heat flux is growing sharply and the curve $q_W = q_W(\tau)$ approaches the vertical asymptote. In the case of nonignition the heat flux decreases and with time it enters into a steady-state mode.

We note that the values of the self-ignition temperature calculated from Eqs. (17) and (23) for magnesium particles in oxygen agree in order of magnitude with the experimental data of [13].

Thus, the mathematical model laid at the basis of the investigation allows one to indicate the conditions under which the steady or nonsteady modes of heat exchange occur in a vessel containing reacting particles for plane, cylindrical, or spherical symmetry. In contrast to [7], we do not use the concept of the coefficient of heat exchange, which, according to [14], is a function of time which is not known in advance for reacting media.

NOTATION

t, time; y, spatial coordinate, measured from center of vessel; T, temperature of disperse medium; T_1, temperature of carrier medium; T_0, temperature of a typical particle; r, coordinate in the equivalent sphere, measured from the particle surface; c, heat capacity; ρ, density; λ_i, coefficient of thermal conductivity (the index 1 refers to the carrier medium and 2 to the particle material); l, half the characteristic dimension of the vessel; r_2, particle radius; E, activation energy of heterogeneous reaction; K_0, pre-exponent; q, thermal effect; k, index of vessel symmetry; λ, coefficient of thermal conductivity of disperse medium; $q_W = -\left.\frac{\partial\theta}{\partial\eta}\right|_{\eta=\pi_l}$, dimensionless heat flux.

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RADIATIVE–CONDUCTIVE HEAT TRANSFER IN TEMPERATURE-WAVE CONDITIONS

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UDC 536.37

Relations describing the radiative–conductive heat transfer in plane temperature-wave conditions are obtained. An analysis is given of the effect of radiative transfer on the measurement of the thermophysical properties of the material by the method of a regular thermal mode of the third kind.

In considering heat-transfer mechanisms in liquids and compressible gases, the role of the radiative transfer is an important and little-studied problem. It is known that heat transfer by radiation may make a notable contribution to the heat conduction of a liquid even at room temperature, and its role increases greatly with increase in temperature [1]. To obtain information on radiative transfer from steady-state experiments, it is necessary to make measurements in cells of different size, which is very troublesome. The potential of nonsteady methods of investigation is fundamentally greater. In [2, 3], the role of radiative heat transfer in experiments on the probing of liquids by heat pulses was investigated; it was shown that in the early stages of this process radiant energy transfer plays a small role, and such experiments allow the pure heat conduction of liquids to be determined. In the present work, the question of radiative heat transfer is investigated in the context of liquid probing by plane temperature waves, which is the main method of measuring the thermal-activity coefficient [1].

A solution is obtained for the problem of the heat transfer in a periodically heated plane layer (represented experimentally as a metal foil) situated in a semitransparent medium. The foil constitutes an infinite plane yz, is situated at the coordinate origin (x = 0), and has a specularly reflecting surface. The heat-transfer equation of the foil in the medium, taking radiation into account, is

$$\frac{W}{s}=\frac{cm}{s}\frac{\partial T_1}{\partial t}-2\lambda\frac{\partial T(0)}{\partial x}+2q(0). \tag{1}$$

The radiant energy flux to an absorbing, nonscattering medium whose optical properties are independent of temperature is given by the relation [4]

$$\begin{aligned} q(x) &= 2(1-R)\,\sigma n^2 T_1^4 E_3(\alpha x) + 2R\sigma n^2\alpha \int_0^\infty T^4(\xi)\,E_2(\alpha(x+\xi))\,d\xi \\ &+ 2\sigma n^2\alpha\left[\int_0^x T^4(\xi)\,E_2(\alpha(x-\xi))\,d\xi - \int_x^\infty T^4(\xi)\,E_2(\alpha(\xi-x))\,d\xi\right], \end{aligned} \tag{2}$$

where $E_3(\alpha x)$ and $E_2(\alpha x)$ are integroexponential functions [5].

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