IGNITION AND SELF-IGNITION OF REACTING SUBSTANCES IN CONDITIONS OF PERFECT THERMAL CONTACT

A. M. Grishin

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The methods of Shvets [1] and of integral relations [2, 3] have been used to solve the problems of ignition and self-ignition of nonvolatile reacting substances, for boundary conditions of the fourth kind.

Consider ignition of a semi-infinite reacting volume of heated medium with outside thermophysical contacts under conditions of perfect thermal contact. According to [4], boundary conditions of the fourth kind are realized for unsteady heat transfer between a solid and a gas or liquid. We shall assume that a reaction of zero order takes place, and that all the thermophysical coefficients are constant. Our problem will be to determine the ignition characteristics. Mathematically the problem reduces to solution of the following system of equations:

$$\frac{\partial^2\theta_1}{\partial x^2} = \frac{\partial \theta_1}{\partial \tau} - \exp\theta_1,\ x > 0, \tag{1}$$

$$\frac{\partial^2\theta_2}{\partial \xi^2} = \frac{\partial \theta_2}{\partial \tau},\ \xi < 0, \tag{2}$$

with boundary and initial conditions

$$\theta_1(0, \tau) = \theta_2(0, \tau),\quad \left.\frac{\partial \theta_1}{\partial x}\right|_{x=0} = n\left.\frac{\partial \theta_2}{\partial \xi}\right|_{\xi=0},$$

$$\theta_1(x, 0) = \theta_1(\infty, \tau) = -\theta_{init},$$

$$\theta_2(\xi, 0) = \theta_2(-\infty, \tau) = 0. \tag{3}$$

In deriving Eq. (1) we made use of the Frank-Kamenetskii transformation [5] for $\exp(-E/RT)$. Equation (1), in accordance with Eq. (6), is a satisfactory description of ignition of condensed reacting substances, even for a reaction of zero order, if $(T_0 - T_{init})c_1\rho_1/q \ll 1$. From the solution of the boundary problem of Eqs. (1) (1)–(3), as a special case, we obtain, when $n \to \infty$, the solution of the problem examined in [7, 8].

In practice, because the chemical reaction rate is an exponential function of temperature, a temperature variation takes place in the vicinity of the interface between the media. It is, appropriate, therefore, to introduce the thermal boundary layer thickness $\Delta_1(\tau)$ and $\Delta_2(\tau)$. Then the boundary condition at $\pm\infty$, and the conditions when $\tau = 0$, take the form

$$\theta_1(\Delta_1, \tau) = -\theta_{init},\quad \theta_2(-\Delta_2, \tau) = 0,$$

$$\Delta_1(0) = \Delta_2(0) = 0. \tag{4}$$

Following Shvets [1], in first approximation we obtain for θ_1 and θ_2,

$$\theta_1^{(1)} = -\frac{\theta_{init}(\Delta_2 + nx)}{\Delta_2 + n\Delta_1},\quad \theta_2^{(1)} = -\frac{\theta_{init}(\Delta_2 + \xi)}{\Delta_2 + n\Delta_1}. \tag{5}$$

Substituting Eq. (5) into the right-hand sides of Eqs. (1) and (2), and integrating the results of the substitution twice with respect to x and ξ, we find the second approximations:

$$\theta_1^{(2)} = = \frac{\dot a_1 x^3}{6} + \frac{\dot b_1 x^2}{2} - \frac{\exp(b_1 + a_1 x)}{a_1^2} + A_1 x + B_1, \tag{6}$$

$$\theta_2^{(2)} = \frac{\dot a_2 \xi^3}{6} + \frac{\dot b_2 \xi^2}{2} + A_2\xi + B_2. \tag{7}$$

The quantities A_1, A_2, B_1, and B_2 are determined from the boundary conditions (3) and (4). Satisfying (6) and (7) by the Shvets conditions [1],

$$\left.\frac{\partial \theta_1}{\partial x}\right|_{x=\Delta_1} = \left.\frac{\partial \theta_2}{\partial \xi}\right|_{\xi=-\Delta_2} = 0, \tag{8}$$

we obtain two ordinary nonlinear differential equations to determine the quantities Δ_1 and Δ_2:

$$\dot a_1\Delta_1^2\left(\frac{\Delta_2}{2} + \frac{n\Delta_1}{3}\right) + \dot b_1\left(\frac{n\Delta_1^2}{2} + \Delta_1\Delta_2 + \frac{n\Delta_2^2}{2}\right) - \frac{n\Delta_2^3\dot a_2}{6} - n\theta_{init} + \frac{(a_1\Delta_2 - n)}{a_1^2}\exp b_1 + \frac{\varepsilon[n - a_1(\Delta_2 + n\Delta_1)]}{a_1^2} = 0, \tag{9}$$

$$\dot a_2\Delta_2^2\left(\frac{\Delta_2}{3} + \frac{n\Delta_1}{2}\right) - \dot b_1\left(\frac{\Delta_1^2}{2} + n\Delta_1\Delta_2 + \frac{\Delta_2^2}{2}\right) - \frac{\dot a_1\Delta_1^2}{2} - \theta_{init} + \frac{\varepsilon - (1 - a_1\Delta_1)\exp b_1}{a_1^2} = 0. \tag{10}$$

If the nonreacting heated medium exhibits a very great heat capacity, the temperature at the boundary between the media remains constant as $n \to \infty$. In this case, instead of the system of equations (9) and (10), we have only one equation remaining, and it may be integrated, taking account of the initial conditions (4), to obtain

$$\Delta_1 = \theta_{init} \times \sqrt{\frac{\theta_{init}}{1 - \varepsilon - \varepsilon\theta_{init}}\left[\exp\frac{6(1 - \varepsilon - \varepsilon\theta_{init})\tau}{\theta_{init}^3} - 1\right]} \approx \sqrt{6\tau}\left(1 + \frac{3\tau}{2\theta_{init}^3}\right). \tag{11}$$

In the general case we solve system (9) and (10) by expanding in series:

$$\Delta_1 = \alpha_1\tau^{1/2} + \beta_1\tau + \delta_1\tau^{3/2} + \ldots,$$

$$\Delta_2 = \alpha_2\tau^{1/2} + \beta_2\tau + \delta_2\tau^{3/2} + \ldots. \quad (12)$$

Substituting Eq. (12) into Eqs. (9) and (10), and equating to zero terms with identical powers of τ, we obtain a system of two nonlinear equations for determining α_1 and α_2, and we obtain systems of linear equations for determining β_1 and β_2 and δ_1 and δ_2. From solution of these equations we find that $\alpha_1 = \alpha_2 = \sqrt{6}$, $\beta_1 = \beta_2 = 0$, and, finally,

$$\delta_1 = \frac{24(1+n)^2(\nu-\varepsilon)p_1}{\sqrt{6}n^2\theta_{\text{init}}^2},$$

$$\delta_2 = \frac{24(1+n)^2(\nu-\varepsilon)p_2}{\sqrt{6}n^2\theta_{\text{init}}^2}. \quad (13)$$

When $n \to \infty$, the first expression in Eq. (12), taking account of the values of α_1, β_1, and δ_1 found, coincides with Eq. (11) to an accuracy including terms containing τ^j, where $j > 3/2$. The first terms in Eqs. (12) represent the role of the boundary layers for nonreacting media. It follows from (6), (7), (11), (12), and (13), that the heat of reaction, for $\theta_{\text{init}} \gg 1$ and moderate values of τ, depends only slightly on the temperature profile and the thickness of the boundary layers. Since Gandin [9] has shown that the sequence of approximations in the Shvets method converges rapidly when solving problems of heat conduction for nonreacting media, and since the perturbation from the heat of reaction is small, higher approximations converge, at least for moderate values of τ. In particular, we find, for small values of τ, from Eq. (6), with the help of Eq. (12), that the temperature at the interface between the media is $-\theta_{\text{init}}/(1+n)$, i.e., it coincides with the corresponding exact value of the temperature of the interface between nonreacting media [10], while the error in the temperature gradient at $x = 0$ does not exceed 8%.

Using the condition $\left.\frac{\partial\theta_1}{\partial x}\right|_{x=0} = 0$, given by Zel'dovich [11], we obtain an equation for the warm-up time of the reacting system:

$$\frac{n\theta_{\text{init}}\Delta_1^2}{2}(\dot{\Delta}_2 + n\dot{\Delta}_1) + n\theta_{\text{init}}\Delta_1(\dot{\Delta}_1\Delta_2 - \Delta_1\dot{\Delta}_2) = \frac{(\Delta_2 + n\Delta_1)^2}{n\theta_{\text{init}}}(\exp b_1 - \varepsilon). \quad (14)$$

Substituting (12) into Eq. (14), and solving the equation with respect to τ, we find an approximate expression for the warm-up time:

$$\tilde{\tau}_* = \frac{n^2\theta_{\text{init}}^2}{4(1+n)^2(\nu-\varepsilon)}. \quad (15)$$

In deriving Eq. (15), we neglected δ_1 and δ_2 in comparison with α_1 and α_2.

It is easy to see from Eq. (15) that when $n \to \infty$, the quantity $\tilde{\tau}_* \to \theta_{\text{init}}^2/4$, and when $n \to 0$, the quantity $\tilde{\tau}_* \to 0$, i.e., $\tilde{\tau}_*$ is a nonmonotonic function of n, and attains a maximum $\tilde{\tau}_* = \tau_{*m}$ for $n = n_*$. The larger is θ_{init}, the sharper and the higher is the maximum value $\tilde{\tau}_*$, and the closer is n_* to 0. For $\theta_{\text{init}} \gg 1$, the quantity $n_* \approx 1/(\theta_{\text{init}} - 2)$. In the limit, as $\theta_{\text{init}} \to \infty$, we obtain $n_* \to 0$ and $\tilde{\tau}_* \to \infty$. The nonmonotonic nature of $\tilde{\tau}_*$ as a function of n is due, evidently, to a specific peculiarity of the Arrhenius function, namely, that the heat liberated from the reaction does not go to zero even at sufficiently high temperatures. This defect in the Arrhenius function was noticed in [12], and the method of sections was used to avoid it in [12]. We shall avoid this defect by means of the method of Spalding [13] and Rosen [14], putting

$$\exp -\frac{E}{RT} \approx C\left(\frac{T - T_{\text{init}}}{T_0 - T_{\text{init}}}\right)^k,$$

$$C = \text{const}, \quad k = \text{const} \gg 1. \quad (16)$$

In an analogous manner, for a heat evolution in the form of Eq. (16), we obtain the warm-up time of the reacting system in the form

$$\tilde{\tau}_{1*} = \frac{k+1}{2}\left(1 + \frac{1}{n}\right)^{k-1}. \quad (17)$$

In this case, with increase of n, the warm-up time monotonically decreases from ∞ at $n = 0$ to $\tilde{\tau}_{1*} = (k+1)/2$ at $n \to \infty$.

For the limiting case $n \to \infty$, we can find a value of warm-up time that is exact within the framework of the approximation of the Shvets method [1]:

$$\tau_* = \frac{\theta_{\text{init}}^3}{6(1-\varepsilon-\varepsilon\theta_{\text{init}})}\ln\frac{2\theta_{\text{init}}(1-\varepsilon)}{(2+\varepsilon)\theta_{\text{init}} - 3(1-\varepsilon)} \approx$$

$$\approx \frac{\theta_{\text{init}}^2}{4} + \frac{3\theta_{\text{init}}}{16} + \frac{3}{32}. \quad (18)$$

Comparing Eq. (18) with the expression for τ_* found in [6] using an electronic computer, we see that its accuracy is quite satisfactory. Thus, for $\theta_{\text{init}} = 5$, 10, 15, 20, 25, 30, with the help of Eq. (18), we have $\tau_* = 7.3$, 27, 59, 104, 161, 231, and from [6] we have $\tau_* = 10$, 30, 60, 100, 150, 210, respectively.

To estimate the accuracy of Eq. (15) for moderate values of n we found τ_* for n = 1 from Eq. (14) by a trial-and-error method. We found $\tau_* = 28.8$ for $\theta_{\text{init}} = 5$, $\tau_* = 957$ for $\theta_{\text{init}} = 10$, and $\tau_* = 24.7 \exp 10$ for $\theta_{\text{init}} = 20$, while from Eq. (15) we have $\tau_* = 20.7$, 934, 25 exp 10, correspondingly. Therfore, the accuracy of Eq. (15) is quite satisfactory within the framework of our approximations.

Knowing τ_*, we can easily find the thickness of the heated layer,

$$\Delta_{1*} = \Delta_1(\tilde{\tau}_*) = \frac{n\theta_{\text{init}}}{1+n}\sqrt{\frac{3}{2(\nu-\varepsilon)}}(1+p_1), \quad (19)$$

the temperature of the interface boundary for $\tau = \tilde{\tau}_*$,

$$\theta_* = \theta_1(0, \tilde{\tau}_*) = -\frac{\theta_{\text{init}}(1+p_2)^2}{[1+p_2+n(1+p_1)]^2} \times$$

$$\times [(1+n)(1+4p_2) + 3p_2(p_2+np_1)], \quad (20)$$

and the amount of heat transferred by the hot medium,

$$Q_* = -\int_0^{\tilde{\tau}_*}\left.\frac{\partial\theta_1^{(2)}}{\partial x}\right|_{x=0} d\tau = \frac{n^2\theta_{\text{init}}^2}{(1+n)^2\sqrt{6(\nu-\varepsilon)}}. \quad (21)$$

Using Eq. (20), with n = 1 and θ_{init} = 5 and 10, we find that $\theta_* = -1.6$ and -2.6, respectively, i.e., in contrast to heat transfer between nonreacting media, the interface temperature in our case increases with increase of τ.

If n decreases from ∞ to 0, it follows from Eq. (21) that Q first increases then decreases to 0. Therefore, there is a maximum value Q_* for $n = n_{1*}$. The right-hand branch of the Q_* curve, corresponding to $n > n_{1*}$, has physical meaning. Thus, for $n > n_{1*}$, to ignite the reagent it is necessary to transfer a greater amount of heat from the hot medium than when $n \to \infty$. This is because the heat evolved in the reaction decreases exponentially with decrease of n.

For $n \to \infty$ we obtain formulas from Eqs. (15) and (21) which are close to the warm-up time and to the quantity Q_*, as found in [6, 8], using a computer.

Thus, a comparison of the limiting cases with the results of machine calculation indicates that the Shvets method [1] converges in our case, at least for $0 < \tau \leq \leq \tau_*$.

For $n \to 0$, in place of Eqs. (19), (20), and (21), we should use formulas which can be obtained in an analogous manner, making use of (16) and (17).

It was shown in [15] that the ignition time can be divided into a warm-up time τ_* and an induction time τ_0. To determine the induction time, we must solve the corresponding problem of self-ignition of a reacting substance, which reduces to solution of the system of equations

$$\frac{\partial \theta_1}{\partial \tau} = \frac{1}{\delta y^i} \frac{\partial}{\partial y}\left(y^i \frac{\partial \theta_1}{\partial y}\right) + \exp \theta_1, \qquad (22)$$

$$\frac{\partial \theta_2}{\partial \tau} = \frac{\sigma}{\delta y^i} \frac{\partial}{\partial y}\left(y^i \frac{\partial \theta_2}{\partial y}\right), \qquad (23)$$

with boundary and initial conditions

$$\left.\frac{\partial \theta_1}{\partial y}\right|_{y=0} = 0, \qquad \theta_1(\tau, 1) = \theta_2(\tau, 1),$$

$$\left.\frac{\partial \theta_1}{\partial y}\right|_{y=1} = \mu \left.\frac{\partial \theta_2}{\partial y}\right|_{y=1},$$

$$\theta_1(0, y) = \theta_2(0, y) = 0. \qquad (24)$$

System (22), (23) with conditions (24) has been solved for i = 2 using a computer [16]. We shall use the method of integral relations to solve it. We first examine the thermal detonation of a reacting sheet. We use the "independent" approximation of nonlinear theory (in the terminology of [3]) assuming that

$$\theta_1 \approx (g - \theta_0) y^2 + \theta_0,$$

$$\exp \theta \approx \exp \theta_0 + (\exp g - \exp \theta_0) y^2. \qquad (25)$$

For the second equation of system (22) and (23) we make the substitution z = y − 1. Then, for $1 \leq y < \infty$, we can use the known solution [10] of the heat conduction equation for a half-space with known thermal flux at its boundary. Using this solution, together with the second and third of conditions (24), we obtain an equation for θ_0 and g:

$$g = \frac{2}{n\sqrt{\pi\delta}} \int_0^\tau \frac{\theta_0(t) - g(t)}{\sqrt{\tau - t}}\, dt. \qquad (26)$$

We obtain a second equation for g and θ_0 by substituting Eq. (25) into Eq. (22) and integrating the result with respect to y from 0 to 1:

$$2\dot{\theta}_0 + \dot{g} = 2\exp\theta_0 + \exp g - \frac{6(\theta_0 - g)}{\delta}. \qquad (27)$$

Therefore, to determine g and θ_0 we have the system (26), (27) with conditions

$$\theta_0(0) = 0, \quad g(0) = 0. \qquad (28)$$

Equation (26) is an Abel equation [17]. Using the transformation formula for this equation [16], we obtain

$$\theta_0 = g + \frac{n}{2}\sqrt{\frac{\delta}{\pi}} \frac{d}{d\tau} \int_0^\tau \frac{g(t)\,dt}{\sqrt{\tau - t}}. \qquad (29)$$

Knowing Eq. (29), we can easily reduce the system (26), (27) to a single equation:

$$g = \frac{1}{3}\int_0^\tau \left\{\exp g(\tau) + 2\exp\left[g + \frac{n}{2}\sqrt{\frac{\delta}{\pi}} \frac{d}{d\tau}\int_0^\tau \frac{g(t)\,dt}{\sqrt{\tau - t}}\right]\right\} d\tau - \frac{n}{\sqrt{\delta\pi}}\int_0^\tau \frac{g(t)\,dt}{\sqrt{\tau - t}} - \frac{n}{3}\sqrt{\frac{\delta}{\pi}} \frac{d}{d\tau}\int_0^\tau \frac{g(t)\,dt}{\sqrt{\tau - t}}. \qquad (30)$$

When $n \to 0$, we have $\theta_0 \to g$ from Eq. (29), i.e., the spatial nonuniformities in the temperature distribution vanish, and we obtain the Semenov-Todes case [18], when there is no heat transmission from the reacting substance. In this case, Eq. (27) can be integrated, and we have

$$\theta_0 = g = -\ln(1 - \tau). \qquad (31)$$

From Eq. (31) we obtain the result that the induction time for n = 0 is equal to 1, in agreement with [18].

If $n \to \infty$, we have the limiting case of Frank-Kamenetskii [5]. In this case, as can be seen from Eq. (26), g = 0, and instead of system (26) and (27), we are left with a single equation which can easily be integrated, and for $\theta_0 \to \infty$ we obtain

$$\tau_0 = \delta \int_0^\infty \frac{d\theta_0}{\delta(1 + 2\exp\theta_0) - 6\theta_0}. \qquad (32)$$

It is easy to see that $\tau_0 < \infty$ and therefore, self-ignition occurs for $\delta > \delta_* = 0.94$, the exact value being $\delta_* = 0.88$ [5]. For $\delta < \delta_*$ there are two steady temperature distributions. The limiting steady value is $\theta_0 = \theta_{0*} = 1.16$.

Thus, for $n \to \infty$ there is a steady temperature distribution and a definite limit, while for $n \to 0$, these do not exist. The question arises as to what occurs at intermediate values. A complete answer to this question can be given by solving Eq. (30), which is a difficult problem, but some indirect data can be obtained without solving Eq. (30). We shall replace $\exp\theta_1$ by 1 in Eq. (22), and apply a Laplace transformation [19] for the linear system obtained with conditions (24). For the transform of θ_1, where θ_1 is the solution of system (22) and (23) with a source equal to 1, we find the expression

$$v_1 = \frac{1}{s^2}\left(1 - \frac{n\,\mathrm{ch}\sqrt{\delta s}\,x}{\mathrm{sh}\sqrt{\delta s} + n\,\mathrm{ch}\sqrt{\delta s}}\right). \quad (33)$$

Our problem is to study the behavior of θ_1 for $\tau \to \infty$. To do this, according to [19], it is sufficient to find the limit of sv_1 as $s \to 0$. It is easy to see that $\lim_{s\to 0} sv_1 = \infty$ and that, therefore, $\lim_{\tau\to\infty}\theta_1 = \infty$, i.e., even for a weak source of heat emission, there is no steady temperature distribution when $\tau \to \infty$. In view of the fact that $\theta_1 > \theta_1$, one of two cases is possible: $\lim_{\tau\to\infty}\theta_1 = \infty$ or $\lim_{\tau\to\tau_0}\theta_1 = \infty$. Therefore, there is no steady temperature distribution for the nonlinear heat source $\exp\theta_1$. This investigation does not resolve the question of the existence of a detonation limit within the framework of the Frank-Kamenetskii approximation [5] for $\exp(-E/RT)$, since it is possible to have a value of δ_*, such that for $\delta > \delta_*$ the limit of $\theta_1(0,\tau)$ when $\tau \to \tau_0$ is equal to ∞. If a detonation limit exists, it depends on n.

For a reacting cylinder, by means of a similar investigation, it is possible to show that again there is no steady temperature distribution.

For a reacting sphere, using the substitution $\varphi = \theta_2 y$ and similar methods, a single integrodifferential equation for g can be obtained. It is easy to verify that for a source equal to 1, a steady temperature distribution does exist here. Since the Arrhenius function $\exp(-E/RT) < 1$ for any T, this conclusion is valid for any values of T. Within the framework of the Frank-Kamenetskii approximation [5], there is evidently a value δ_*, such that for $\delta > \delta_*$ there is nonsteady temperature distribution.

NOTATION

$\theta = (T - T_0)E/RT_0^2$ is the dimensionless temperature; $x = r((k_0E/\lambda_1RT_0^2)\exp(-E/RT))^{1/2}$ is the dimensionless coordinate; $\tau = (qk_0Et/C_1\rho_1RT_0^2)\exp(-E/RT_0)$ is dimensionless time; T_0 is the initial temperature of a heated nonreacting medium, and also the initial temperature of the reagent for the self-ignition problem; T_{init} is the initial temperature of the reagent in the ignition problem; R is the universal gas constant; q is the thermal effect of the reaction; r is a dimensional coordinate; k_0 is a preexponential factor; E is activation energy; λ is the thermal conductivity; ρ is density; c is heat capacity; t is time; $\varkappa$ is thermal diffusivity; subscripts 1 and 2 correspond to the reacting substance and the nonreacting medium, respectively; $\varepsilon = \exp(-\theta_{init})$; $\nu = \exp -\theta_{init}/(1+n)$; $\theta_{init} = (T_0 - T_{init}E)/RT_0^2$; $n = (\lambda_2\rho_2c_2/\lambda_1\rho_1c_1)^{1/2}$ is the relative thermal activity coefficient for the known reaction medium; $a_1 = -\theta_{init}/(\Delta_2 + n\Delta_1)$; $b_1 = b_2 = -\theta_{init}\Delta_2/(\Delta_2 + n\Delta_1)$; $a_2 = -\theta_{init}/(\Delta_2 + n\Delta_1)$; $A_1 = (\exp b_1/a_1) + nA_2$; $B_1 = (\exp b_1/a_1^2) + B_2$; $A_2 = (\Delta_2 + n\Delta_1)^{-1}[(\varepsilon/a_1^2) + (b_1/2)\cdot(\Delta_2^2 - \Delta_1^2) - (\dot a_2\Delta_2^3 + \dot a_1\Delta_1^3)/6 - \theta_{init} - (1 + a_1\Delta_1/a_1^2)\times\exp b_2]$; a dot above a symbol indicates differentiation with respect to τ; $p_1 = ((9 + 9n + 4\theta_{init})(\nu - \varepsilon) - 9n\times\varepsilon\theta_{init})/24(1+n)(\nu - \varepsilon)\theta_{init}$; $p_2 = (9(1+n)(\nu - \varepsilon) - n\theta_{init}(6\nu + 5\varepsilon))/24(1+n)(\nu - \varepsilon)\theta_{init}$; $\tau_1 = (k_0Ct/c_1)\times p_1(T_0 - T_{init})$ is dimensionless time; $\xi = x/\sqrt{\sigma}$ is a dimensionless coordinate; $y = r/r_0$ is the dimensionless ambient radius; $\mu = \lambda_2/\lambda_1$; $\sigma = \varkappa_2/\varkappa_1$; r_0 is the characteristic dimension of the reacting volume; i = 0, 1, 2 refer to the plate, cylinder, and sphere, respectively; $\delta = (qk_0Er_0^2/\lambda_1RT_0^2)\exp(-E/RT_0)$ is the Frank-Kamenetskii parameter [5].

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