A MODEL FOR THE PERMANENT TRANSIENT STATE IN THE COMBUSTION ZONE FOR A MIXED CONDENSED SYSTEM

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Previous studies [1,2] have examined the features of the combustion mechanisms of condensed mixtures arising from the heterogenous random structure of the condensed phase. Although the mean burning rate remains constant for a given specimen if the components are adequately mixed and if the temperature and pressure are constant, the microstructure of the combustion zone fluctuates. Either fuel or oxidizing agent may appear at a given point in a coordinate system linked to the mean level of the burning surface. The two components burn at different rates even in the steady state, so the instantaneous rate of combustion varies, and the surface of the condensed phase is not planar. If the oxidant is ammonium perchlorate, it can burn on its own [3], whereas patches of fuel can burn only when oxidant is supplied (usually by molecular diffusion). It is also possible for parts rich in fuel to burn independently and to have a steady burning rate dependent on the external conditions, e.g., bidisperse systems [4] or oxidant and fuel floating on the burning surface.

Figure 1 shows schematically the condensed phase and burning surface. Along the x-axis lie in succession areas of fuel (shaded area, 2) and oxidant (white, 1). To calculate the mean speed of the burning surface along the x-axis we need to derive an averaged solution to a complicated problem of uneven thermal conduction, diffusion, and reaction in a system with random component distributions. This is essentially a statistical problem, since even regular (e.g., spherical) particles of oxidant in general are randomly distributed.

In view of the complexity, it is of interest to examine simpler models, e.g., a stratified condensed system in which the properties vary only along the perpendicular to the combustion front (Fig. 2).

Let $\bar{u}_1$ and $\bar{u}_2$ be the steady-state burning rates for substances 1 and 2 (it is assumed that both can burn independently). An example is alternating layers of different grades of gunpowder.

If the thicknesses h_1 and h_2 are fairly large, the mean rate can be calculated roughly as

$$\bar{u} = \bar{u}_1\bar{u}_2\frac{h_1 + h_2}{\bar{u}_1 h_1 + \bar{u}_2 h_2}.$$

This simple formula is not correct in the general case because the front speeds in the system of Fig. 2 will not be $\bar{u}_1$ in 1 and $\bar{u}_2$ in 2; when the front passes through the 1-2 boundary there will be a transitional state between $\bar{u}_1$ and $\bar{u}_2$. Zel'dovich's theory [5] of burning for low and high explosives indicates that the transients in a condensed system are substantially dependent on the structure of the heated layer. Zel'dovich [6] has examined the combustion of individual thin plates of low explosive. If the thickness of a plate becomes comparable with the effective thickness of the heated layer, which is $x_T = \varkappa/\bar{u}$, where $\varkappa$ is the thermal diffusivity, then the rate of combustion will differ appreciably from the steady-state value.

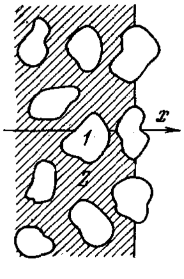

Fig. 1

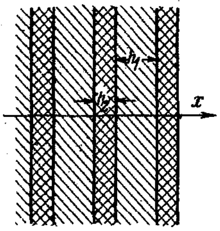

Fig. 2

Near the 1-2 contact there is a transition zone whose thickness is several x_T. It is not always possible for combustion to pass from 1 to 2. The theory of ignition for solid rocket fuels [7,8] indicates that necessary conditions for ignition are that the surface attains a temperature T_s and that the temperature gradient does not exceed a certain critical value. Substance 2 will ignite only if $T_{s1} > T_{s2}$ and the temperature gradient at the boundary is below the critical value. Clearly, T_{s2} will be reached at the interface before the gasification front in substance 1 reaches the interface. If the gradient is then less than critical, 2 may ignite before 1 has burned up completely. This mechanism may be responsible for additional dispersal of the condensed phase. The usual dispersal mechanism [9] involves a vigorous exothermic reaction in the condensed phase, whereas the above mechanism can occur in a chemically inert but heterogeneous condensed medium.

The burning front cannot propagate in a one-dimensional stratified system in the absence of heat relase in the condensed phase if $T_{s1} \neq T_{s2}$, since the burning will die out on passage from 2 to 1 ($T_{s1} > T_{s2}$). In an actual system, this does not happen for the following reasons: a) the actual structure of the condensed phase is not one-dimensional, which provides supply of heat and is the lacking component to adjacent parts of the burning surface, b) in most cases the burning occurs with appreciable heat release in the condensed phase [9], so the ignition-extinction conditions are different from those derived in Zel'dovich's theory. It is therefore of interest to examine by experiment the burning characteristics of a one-dimensional layered system with layers differing substantially in burning rate not only as regards simulation of a persistent transient zone but also as an example of combustion in a system in which the very possibility of combustion is determined by reaction in the condensed phase. Even if substances 1 and 2 have similar steady-state burning rates, the burning zone is a permanently transient one if the thermophysical characteristics of the substances are sufficiently different. An example is provided by metallized and unmetallized ballistic powders: the two burn similarly but the thermal conductivities can differ substantially. The condition for the effect to be observable is that the layer thicknesses h should be comparable with the characteristic size of the heated layers in 1 and 2; if this is so, the burning rate will always be nonstationary. We take the thermophysical parameters of the condensed phase as $\lambda = 5 \cdot 10^{-4}$ cal/cm-sec-deg, $\rho = 1.5$ g/cm^3, and $c_p = 0.33$ cal/g-deg, which give for burning rates of 1 and 10 mm/sec that $x_{T1} = 0.1$ mm, $x_{T2} = 0.01$ mm. Although there is serious difficulty in actually making a system with a characteristic layer thickness of 0.1 mm, this difficulty can, in principle, be overcome. The characteristic particle sizes in real mixtures are 10–100 μ, so the nonstationary effect does play a substantial part.

A direct result of this nonstationary combustion is that the two-temperatures assumption [10] is not applicable to burning of a real condensed system of random structure, since the basic assumption (equal gasification rates for oxidant and fuel) may not be met under real conditions.

Transient conduction in the burning zone does not exhaust the causes of nonstationary burning in mixtures, as a major part is played by transient heat transfer in directions perpendicular to the vector for the mean velocity of propagation of the combustion front. The literature [11] discusses only diffusion mixing of components in the combustion zone, whereas transverse heat transfer is equally important.

Consider a simple example of combustion in a layered system. We assume that the two fuels are identical in characteristics apart from the thermal diffusivity and that this quantity changes smoothly in the following way:

$$\varkappa(x) = \varkappa_0 + \varkappa_1 \sin kx. \tag{1}$$

We assume also that $\varkappa_1/\varkappa_0$ is small, and so we can use Novozhilov's method for calculating the mean burning rate under conditions of varying thermal conduction in accordance with Zel'dovich's theory of transient-state burning [5].

In the case of burning with a harmonically varying external pressure, the following is the equation for thermal conduction in the condensed phase as written in the laboratory coordinate system:

$$\frac{\partial T}{\partial t} = \frac{\partial}{\partial x}\left[\varkappa(x)\frac{\partial T}{\partial x}\right]. \tag{2}$$

The condensed phase lies in the volume

$$x \leqslant x_s = \int_0^t u(\eta)\,d\eta.$$

The boundary conditions for (2) express the constancy of the temperatures at the surface and at depth in the condensed phase:

$$T(x_s) = T_s, \qquad T(-\infty) = T_0. \tag{3}$$

Constancy of $T(x_s)$ means that the combustion of the condensed system is described by the Zel'dovich-Belyaev model [5].

The nonstationary burning rate u(t) [5, 13] is a function of the pressure and the temperature gradient at the surface of the condensed phase:

$$u(t) = \Phi(p, (\partial T/\partial x)x_s). \tag{4}$$

We assume that the following formula describes the steady-state dependence of the combustion rate on the pressure and initial temperature:

$$\bar{u} = f(p)e^{\beta T_0}. \tag{5}$$

Then [5,13] the nonstationary burning law of (4) is derived by expressing the initial temperature in (5) via the temperature gradient at the burning surface:

$$T_0 = T_s - \frac{\varkappa}{u}\left(\frac{\partial T}{\partial x}\right)_s. \tag{6}$$

We substitute (6) into (5) to relate the nonstationary burning rate to the temperature gradient at the burning surface:

$$u = f(p)\exp\left(\beta T_s - \frac{\varkappa}{u}\beta\left(\frac{\partial T}{\partial x}\right)_s\right). \tag{7}$$

It is convenient to transfer to a moving coordinate system linked to the burning surface:

$$x' = x + \int_0^t u(\eta)\,d\eta, \quad t = t'.$$

Here (2), (3), and (7) become

$$\frac{\partial T}{\partial t'} + u(t')\frac{\partial T}{\partial x'} = \frac{\partial}{\partial x'}\left[\varkappa(x', t')\frac{\partial T}{\partial x}\right], \quad x' \leqslant 0,\ t' > 0, \tag{8}$$

$$x_s' = 0. \quad T'_{x'=0} = T_s, \quad T(-\infty) = T_0, \tag{9}$$

$$u = f(p)\exp\left[\beta T_s - \frac{\varkappa}{u}\beta\left(\frac{\partial T}{\partial x'}\right)_s\right]. \tag{10}$$

All the subsequent discussion relates to this coordinate system, and the primes are omitted for simplicity. The following formula defines the unperturbed temperature distribution, which corresponds to $\varkappa_1 = 0$:

$$\overline{T}(x) = T_0 + (T_s - T_0)\exp((\bar{u}/\varkappa_0)x), \tag{11}$$

in which $\bar{u}$ is the steady-state burning rate for $\varkappa_1 = 0$. In the dimensionless variables

$$\xi = \frac{\bar{u}}{\varkappa_0}x, \quad \tau = \frac{u^2}{\varkappa}t,$$

$$\vartheta = \frac{T - T_0}{T_s - T_0}, \quad v = \frac{u}{\bar{u}}, \quad \Omega = k\frac{\varkappa_0}{u},$$

we replace (1) and (5)-(8) by

$$\frac{\varkappa}{\varkappa_0} = 1 + \frac{\varkappa_1}{\varkappa_0}\sin\left[\Omega\left(\xi - \int_0^\tau v(\eta)\,d\eta\right)\right], \tag{12}$$

$$\frac{\partial\vartheta}{\partial\tau} + v(\tau)\frac{\partial\vartheta}{\partial\xi} = \frac{\partial}{\partial\xi}\left[\frac{\varkappa(\xi,\tau)}{\varkappa_0}\frac{\partial\vartheta}{\partial\xi}\right] \quad (\xi \leqslant 0), \tag{13}$$

$$\vartheta(0) = 1, \quad \vartheta(-\infty) = 0, \quad \overline{\vartheta} = e^{\xi}, \tag{14}$$

$$v\bar{u} = f(p)\exp\left[\beta T_s - \frac{\varkappa\varepsilon}{\varkappa_0 v} - \left(\frac{\partial\vartheta}{\partial\xi}\right)_0\right], \quad (\varepsilon \equiv \beta(T_s - T_0)). \tag{15}$$

As $\varkappa_1/\varkappa_0 \ll 1$, we use successive approximation to solve (12)-(15) by putting the solution as

$$\vartheta(\xi, \tau) = \overline{\vartheta}(\xi) + \delta\vartheta^{(1)}(\xi, \tau) + \delta\vartheta^{(2)}(\xi, \tau) + \ldots,$$

$$v(\tau) = 1 + \delta v^{(1)}(\tau) + \delta v^2(\tau) + \ldots,$$

$$\delta\vartheta^{(1)}, \quad \delta\vartheta^{(1)} \sim \varkappa_1/\varkappa_0, \quad \delta\vartheta^{(2)}, \quad \delta v^{(2)} \sim (\varkappa_1/x_0)^2. \tag{16}$$

Here $\delta\vartheta^{(1)}$ and $\delta v^{(1)}$ are linear perturbations to the steady-state solution. In the linear approximation, the perturbations are harmonic functions of time having the dimensionless frequency Ω, while the mean burning rate remains unchanged. In order to calculate the change in the mean rate we have to consider higher (nonlinear) approximations, since we expect the sum and difference frequencies 2Ω and 0 to appear in the second approximation; i.e., we expect a constant component. We derive the solutions in the first and second approximations. We substitute (16) into (12)-(15) and neglect terms whose order of smallness is higher than the first to find the equations and conditions for $\delta\vartheta^{(1)}$ and $\delta v^{(1)}$, which can be written as

$$\frac{\partial^2\delta\vartheta^{(1)}}{\partial\xi^2} - \frac{\partial\delta\vartheta^{(1)}}{\partial\xi} - \frac{\partial\delta\vartheta^{(1)}}{\partial\tau} =$$

$$= \delta v^{(1)}e^{\xi} - \frac{\varkappa_1}{\varkappa_0}\frac{\partial}{\partial\xi}\left[e^{\xi}\sin\Omega(\xi - \tau)\right], \tag{17}$$

$$\delta\vartheta^{(1)}(0, \tau) = \delta\vartheta^{(1)}(-\infty, \tau) = 0, \tag{18}$$

$$g\delta v^{(1)} = \left(\frac{\partial\delta\vartheta^{(1)}}{\partial\xi}\right)_0 - \frac{\varkappa_1}{\varkappa_0}\sin\Omega\tau \qquad \left(g = \frac{\varepsilon - 1}{\varepsilon}\right). \tag{19}$$

It is best to seek the solution to (17)–(19) via complex representation of $\delta\vartheta^{(1)}$ and $\delta v^{(1)}$:

$$\delta v^{(1)} = \theta(\xi)\, e^{i\Omega\tau}, \qquad \delta v^{(1)} = V e^{i\Omega\tau}, \tag{20}$$

in which $\theta(\xi)$ and V are complex amplitudes. The linear perturbations in the temperature and velocity are determined by the real parts of (20).

We get from (17)–(19) for the complex amplitudes that

$$\frac{d^2\theta}{d\xi^2} - \frac{d\theta}{d\xi} - i\Omega\theta = Ve^{\xi} - i\frac{\varkappa_1}{\varkappa_0}(1-\Omega)\, e^{(1-i\Omega)\xi}, \tag{21}$$

$$\theta(0) = \theta(-\infty) = 0, \tag{22}$$

$$gV = i\varkappa_1/\varkappa_0 + (d\vartheta/d\xi)_0. \tag{23}$$

The solution to (21) subject to (22) is

$$\theta(\xi) = \frac{V}{i\Omega}(e^{\alpha_1\xi} - e^{\xi}) + \frac{\varkappa_1}{i\Omega\varkappa_0}\frac{1-i\Omega}{\Omega+2i}(e^{\alpha_1\xi} - e^{-i\Omega\xi+\xi}),$$

$$\alpha_1 = {}^1/_2\,(1 + \sqrt{1+4i\Omega}). \tag{24}$$

We differentiate (24) and use (23) to get an explicit expression for the amplitude of the linear speed perturbation:

$$V = i\,\frac{\varkappa_1}{\varkappa_0}\frac{\alpha_1 - 1}{(\alpha_1 g - 1)(2-\alpha_1)}. \tag{25}$$

Consider now the second approximation. We substitute (16) into (12)–(15) and retain terms of the second order of smallness to get

$$\frac{\partial\delta\vartheta^{(2)}}{\partial\xi^2} - \frac{\partial\delta\vartheta^{(2)}}{\partial\xi} - \frac{\partial\delta\vartheta^{(2)}}{\partial\tau} = \delta v^{(2)} e^{\xi} + \delta v^{(1)}\frac{\partial\delta\vartheta^{(1)}}{\partial\xi} +$$

$$+ \frac{\varkappa_1}{\varkappa_0}\Omega\frac{\partial}{\partial\xi}\left[e^{\xi}\cos\Omega(\xi-\tau)\int_0^{\tau}\delta v^{(1)}d\tau\right] -$$

$$- \frac{\varkappa_1}{\varkappa_0}\frac{\partial}{\partial\xi}\left[\frac{\partial\delta\vartheta^{(1)}}{\partial\xi}\sin\Omega(\xi-\tau)\right], \tag{26}$$

$$\delta\vartheta^{(2)}(0) = \delta\vartheta^{(2)}(-\infty) = 0, \tag{27}$$

$$g\delta v^{(2)} = \left(\frac{\partial\delta\vartheta^{(2)}}{\partial\xi}\right)_0 + \frac{2\varepsilon-1}{2\varepsilon}(\delta v^{(1)})^2 - \delta v^{(1)}\left(\frac{\partial\delta\vartheta^{(1)}}{\partial\xi}\right)_0 +$$

$$+ \left(\delta v^{(1)} - \left(\frac{\partial\delta\vartheta^{(1)}}{\partial\xi}\right)_0\right) - \frac{\varkappa_1}{\varkappa_0}\Omega\int_0^{\tau}\delta v^{(1)}\,d\eta\cos\Omega\tau. \tag{28}$$

As we are interested only in the constant component of the rate perturbation, there is no need to solve (26)–(28) completely.

Let $\delta\vartheta_c^{(2)}$ and $\delta v_c^{(2)}$ be the constant components of the temperature and rate perturbations, which satisfy (26)–(28), in which we put $\partial/\partial t = 0$ and discard time-dependent terms. Then the equation for δv_c, which does not contain an unknown function on the right, is integrated directly between the limits $\xi = 0$, $\xi = -\infty$ with allowance for (27) to give an expression for the constant component of the temperature perturbation at the burning surface:

$$\left(\frac{d\delta\vartheta^{(2)}}{d\xi}\right)_0 = \delta v_c^{(2)} + \frac{|V|(1-g)}{2}\frac{\varkappa_1}{\varkappa_0}\sin\alpha_V + \frac{1}{2}\left(\frac{\varkappa_1}{\varkappa_0}\right)^2. \tag{29}$$

In (29) we have used the representation $V = |V|e^{i\alpha_V}$ for the complex amplitude of the linear rate perturbation.

Equation (28) can be written as follows after the time-dependent terms have been discarded:

$$g\delta v_c^{(2)} = \left(\frac{d\delta\vartheta_c{}^2}{d\xi}\right)_0 + \frac{(1-g)}{4}|V|^2 -$$

$$- \frac{|V|(1-g)}{2}\frac{\varkappa_1}{\varkappa_0}\sin\alpha_V - \frac{1}{2}\left(\frac{\varkappa_1}{\varkappa_0}\right)^2. \tag{30}$$

From (29) and (30) we get that

$$\delta v_c^{(2)} = -\frac{|V|^2}{4}, \quad V = i\,\frac{\varkappa_1}{\varkappa_0}\frac{\alpha_1 - 1}{(\alpha_1 g - 1)(2-\alpha_1)}. \tag{31}$$

From (31) we can consider the limiting cases in which 1) $\varkappa_0/\bar{u}$ is much greater than $1/k$ (characteristic length for variation in the thermal diffusivity), i.e., $\Omega \gg 1$, and 2) $\Omega \ll 1$:

$$\delta v_c^{(1)} \approx -\frac{1}{4}\frac{\varkappa_1}{\varkappa_0 g\sqrt{\Omega}} \quad (\Omega \gg 1),$$

$$\delta v_c^{(2)} \approx -\frac{\varkappa_1}{\varkappa_0}\frac{\Omega}{4|g-1|}\;(\Omega \ll 1). \tag{32}$$

It follows from (31) and (32) that the burning rate for a system with the variable thermal conductivity of (1) is less than that for $\varkappa_1 = 0$ and that $\bar{u}$ is not affected by harmonic perturbations in the thermal conductivity whose period is much less or much greater than the characteristic size of the thermal layer.

The above analysis can be applied in a straightforward way to burning with a variable temperature at the burning surface [12,14]. Analogy with weak harmonic pressure perturbations indicates that periodic variation in the parameters of the condensed phase should result in resonant modes of combustion, in which the mean rate for a layered system will substantially exceed the mean steady-state rates for both components.

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