TWO-DIMENSIONAL STABILITY OF THE COMBUSTION OF CONDENSED SYSTEMS

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The question of the combustion stability of condensed systems relative to curvature of the front is investigated in a linear approximation. Two of the simplest combustion models are examined, a gasless system and a model of flameless combustion of a solid fuel. In the first case, the combustion products are condensed, just as are the initial materials, and in the second the solid fuel is converted into a gas in which no chemical reactions occur. Boundaries of the stability of the stationary combustion mode are found. It is shown that gasless systems are less stable with respect to two-dimensional perturbations than to one-dimensional perturbations. For the flameless combustion model the result depends on the relationship between the thermophysical constants of the initial material and the products. The question of the influence of heat emission on the one-dimensional stability of the gasless composites is considered. An increase in the heat emission diminishes the stable combustion region, where a one-dimensional instability originates earlier than collapse of combustion occurs because of strong heat emission to the wall.

1. Stability of the Combustion of Gasless Systems

Combustion of a solid fuel is ordinarily accompanied by the transition of the material from the condensed into the gaseous state. However, there are cases when reaction products remain in the condensed state behind the front of an exothermal reaction being propagated over a solid fuel. An example might be the combustion of thermites which were investigated experimentally in [1,2]. An analogous picture originates in the propagation of a polymerization front in a condensed medium. The question of the stationary velocity of reaction front propagation in such systems was considered theoretically in [3-6].

Let us investigate the combustion stability of a gasless composition by using the assumption of a strong temperature dependence of the chemical reaction rate. This assumption permits consideration of the narrow zone of initial material conversion in comparison with the width of the heating zone, and to consider it as a surface separating the initial material (zone 1) and the reaction products (zone 2). (The quantities referring to each of these zones are marked with the subscripts 1 and 2, respectively.) Let us take a coordinate system in which the unperturbed powder surface is at rest (x = 0) and the initial material located at x < 0 moves at the velocity u equal to the stationary propagation velocity of the chemical reaction front.

The stationary temperature distributions

$$T_1^0 = T_0 + (T_a - T_0) e^{ux/x}, \qquad T_2^0 = T_a$$
(1.1)

satisfy the same heat-conduction equation

$$\lambda \frac{d^2 T^0}{dx^2} - \rho u c \frac{d T^0}{dx} = 0 \tag{1.2}$$

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© 1974 Consultants Bureau, a division of Plenum Publishing Corporation, 227 West 17th Street, New York, N. Y. 10011. No part of this publication may be reproduced, stored in a retrieval system, or transmitted, in any form or by any means, electronic, mechanical, photocopying, microfilming, recording or otherwise, without written permission of the publisher. A copy of this article is available from the publisher for \$15.00. and the boundary conditions

$$T_{1^{0}}|_{x=-\infty} = T_{0}, \qquad T_{1^{0}}|_{x=0} = T_{a}$$

$$T_{2^{0}}|_{x=0} = T_{a}, \qquad \frac{dT_{2}^{0}}{dx}|_{x=\infty} = 0$$
(1.3)

Here T_0 is the initial powder temperature, $T_a = T_0 + q/c$ is the combustion temperature, ρ is the density, c is the specific heat, $\varkappa = \lambda/\rho c$ is the coefficient of temperature conductivity (λ is the heat-conduction coefficient), and q is the thermal effect of the reaction. The stationary temperature values are marked with degrees.

Using the method of small perturbations, let us give the curvature of the combustion surface in the following manner:

$$\xi = D \exp\left(\omega t + iKy\right) \tag{1.4}$$

where y is a coordinate directed along the unperturbed reaction front, ω is the frequency, t is the time, K is the wave number, and D is the amplitude of the perturbation. Without limiting the generality, the two-dimensional problem can be examined.

Let us seek the perturbed solution in the form

$$T_{i} = T_{i}^{0} + T_{i}'(x) \exp(\omega t + iKy) \qquad (i = 1, 2)$$
(1.5)

Substituting (1.5) into the nonstationary heat-conduction equation

$$\rho c \, \frac{\partial T_i}{\partial t} = \lambda \left(\frac{\partial^2 T_i}{\partial x^2} + \frac{\partial^2 T_i}{\partial y^2} \right) - \rho u c \, \frac{\partial T_i}{\partial x} \tag{1.6}$$

results in ordinary second-order differential equations in $T_1'(x)$, $T_2'(x)$, whose solution yields

$$T_{1}' = A \exp(z_{1}ux / 2\varkappa), \qquad z_{1} = 1 + \sqrt{1 + 4\Omega + s^{2}}$$

$$T_{2}' = B \exp(z_{2}ux / 2\varkappa), \qquad z_{2} = 1 - \sqrt{1 + 4\Omega + s^{2}}$$

$$\Omega = \frac{\varkappa}{z^{2}}\omega, \quad s = 2 \frac{\varkappa}{z} K$$
(1.7)

Here Ω is the dimensionless frequency, s characterizes the ratio between the width of the Michelson heating zone \varkappa/u and the perturbation wavelength $2\pi/K$, and A and B are integration constants. The condition that the perturbations damp out at infinity was used in selecting the signs of z_1 and z_2 .

The perturbed solutions are interrelated by conditions on the surface where the chemical reaction occurs: the temperature is continuous, but the heat flux is changed because of heat liberation in the reaction zone. In the approximation under consideration we have

$$x = \xi, \quad T_1 = T_2, \quad -\lambda \frac{\partial T_1}{\partial x} + \rho \left(u - \frac{\partial \xi}{\partial t} \right) q = -\lambda \frac{\partial T_2}{\partial x}$$
 (1.8)

It has here been taken into account that the derivative with respect to the normal is different from the derivative with respect to x by a second-order infinitesimal.

Henceforth, let us consider the combustion velocity to depend only on the temperature in the zone of chemical conversion. Then in the linear approximation the change in combustion velocity with temperature can be described by the coefficient

$$k = (T_a - T_0) \frac{d \ln u}{dT_0}$$
(1.9)

whose explicit expression depends on the form of the stationary combustion law $u(T_a)$. In the case of the Arrhenius chemical reaction rate dependence, which was considered in [3-6], we have when we neglect the power dependence as compared to the exponential dependence

$$u \sim \exp\left(-E/2RT_{a}\right), \quad k = \frac{(T_{a} - T_{0})E}{2RT_{a}^{2}}$$
 (1.10)

Here E is the activation energy, and R is the gas constant.

The introduction of a coefficient of temperature response of the combustion velocity permits writing the condition of the total consumption of the reacting material as follows:

$$x = \xi, \quad \frac{\partial \xi}{\partial t} = -\frac{ku(T_2 - T_0)}{T_a - T_0} \tag{1.11}$$

Let us linearize (1.8), (1.11) relative to small admixtures; hence for x = 0 we have

$$\frac{dT_1^{\circ}}{dx}\xi + T_1' = \frac{dT_2^{\circ}}{dx}\xi + T_2'$$

$$\lambda \frac{d^2T_1^{\circ}}{dx^2}\xi + \lambda \frac{\partial T_1'}{\partial x} + \rho \frac{\partial \xi}{\partial t}q = \lambda \frac{d^2T_2^{\circ}}{dx^2}\xi + \lambda \frac{\partial T_2'}{\partial x}$$

$$\frac{\partial \xi}{\partial t} = -\frac{k}{T_a - T_0} \left(\frac{dT_2^{\circ}}{dx}\xi + T_2'\right)u \qquad (1.12)$$

Substituting the solutions (1.1), (1.4), and (1.7) into these relations, we obtain a homogeneous system of three linear equations in A, B, and D, whose solvability condition yields

 $16\Omega^{3} + 4\Omega^{2} (1 + 4k - k^{2} + s^{2}) + 4\Omega k (1 + s^{2}) + s^{2}k^{2} = 0$ (1.13)

Setting $\Omega = \pm i\psi$ here, we find the stability boundary $k_1(s^2)$

$$k_1 = (4 + 3s^2 + \sqrt{(4 + 3s^2)^2 + 4(1 + s^2)^3})/2(1 + s^2)$$
(1.14)

The minimal value is $k_1 = 4$ and is achieved for s = 1. Putting s = 0 into (1.14), we obtain that the domain of instability to one-dimensional perturbations is located at $k > 2 + \sqrt{5} \approx 4.24$. Therefore, the chemical reaction front is more stable relative to one-dimensional, than to three-dimensional perturbations.

The frequency is pure imaginary on the stability boundary, i.e., the loss of stability is oscillatory in nature. It is expressed as a function of the perturbation wavelength in the form

$$\psi^2 = \frac{1}{8} \left(4 + 3s^2 + \sqrt{(4 + 3s^2)^2 + 4(1 + s^2)^3} \right)$$
(1.15)

As we recede from the stability boundary (an increase in k), we can reach a domain where the perturbations will grow exponentially. The boundary of the oscillatory and exponential instability $k_2(s^2)$ can be obtained by using Cardano formulas for the cubic equation (1.13). It is determined by the equation

$$(s^{2})^{3} - (s^{2})^{2} (2k_{2}^{2} + 6k_{2} - 3) + s^{2} (k_{2}^{4} - 10k_{2}^{3} + 26k_{2}^{2} - 12k_{2} + 3) + k_{2}^{2} - 6k_{2} + 1 = 0$$
(1.16)

This curve can be constructed numerically by finding the zeroes of the function in the left side of (1.16) for each k. In particular, for s = 0 we have $k_2 = 3 + 2\sqrt{2} \approx 5.83$. Graphs of the functions $k_1(s^2)$, $k_2(s^2)$, and $\psi(s^2)$ are represented in Fig. 1. The instability domain is located above the curve $k_1(s^2)$.

The stability analysis made above shows that gasless systems are less stable to two-dimensional than to one-dimensional perturbations. The loss of one-dimensional stability occurs in the domain of developed two-dimensional perturbations. This circumstance should be taken into account in attempts at a numerical computation of the nonstationary velocity of the chemical reaction front in an unstable domain. The onedimensional approach to such problems is incorrect.

It should also be noted that the stability analysis performed above duplicates the investigation of the diffusion-thermal stability of a gas flame to three-dimensional perturbations [7] to a significant degree. The authors of [7] limited themselves to a clarification of the influence of the factor $D - \kappa$ (D is the diffusion coefficient) on the stability of a gas flame to longwave perturbations. Moreover, the problem was solved for a medium with constant density. Hence, if we do not limit ourselves to just longwave perturbations and we pass to the limit as $D/\kappa \rightarrow 0$ in the appropriate relations [7] then we obtain the dispersion relation (1.13) which yields unstable roots for k > 4.



2. Influence of Heat Emission on the One-Dimensional Instability of Gasless Compositions

Ya. B. Zel'dovich [8] has shown that heat emission can result in the collapse of combustion. A minimal temperature in the reaction zone T_b^* at which stationary combustion is still possible was found in [8] and the combustion velocity corresponding to this temperature u_b^* has been calculated

$$T_{b}^{*} = T_{a} - \frac{RT_{a}^{2}}{E}, \quad u_{b}^{*} = \frac{u}{\sqrt{e}}$$
 (2.1)

Although the theory of combustion limits has been developed for gases in [8], it can be applied also to the combustion of condensed systems subjected to an Arrhenius kinetics [the combustion law is expressed as a dependence of the form (1.10) in this case].

It is shown in this section that the instability of the stationary combustion mode of a gasless compound originates earlier (with fewer heat losses) than col-

lapse of combustion is realized. To do this, let us investigate the one-dimensional stability of a reaction front being propagated under heat-emission conditions. Considering the temperature constant over the tube cross section, let us add a term taking account of the heat losses to the right side of (1.6).

$$\rho c \, \frac{\partial \Gamma_i}{\partial t} = \lambda \frac{\partial^2 \Gamma_i}{\partial x^2} - \rho u c \, \frac{\partial \Gamma_i}{\partial x} - \delta \left(T_i - T_0 \right) \tag{2.2}$$

Here δ is the heat-transfer coefficient.

Let us first present the stationary relations. In place of (1.1) we have

$$T_{1}^{\circ} = T_{0} + (T_{b} - T_{0}) \exp \frac{u_{b} (1 + \beta) x}{2\kappa} T_{2}^{\circ} = T_{0} + (T_{b} - T_{0}) \exp \frac{u_{b} (1 - \beta) x}{2\kappa}$$
 (β² = 1 + 4 $\frac{\delta}{\lambda} \frac{x^{2}}{u_{b}^{2}}$) (2.3)

Here T_b and u_b are the temperature in the reaction zone and the linear velocity of combustion under heat-emission conditions, respectively, β is a parameter characterizing the intensity of heat emission ($\beta = 1$, $T_b = T_a$, $u_b = u$ for $\delta = 0$).

The equation of thermal energy balance on the combustion surface

$$x = 0, \quad -\lambda \frac{dT_1^{\circ}}{dx} + \rho u_b q = -\lambda \frac{dT_2^{\circ}}{dx}$$
(2.4)

determines the combustion temperature

$$T_{b} = T_{0} + \frac{q}{c\beta} \approx \frac{T_{a}}{\beta}$$
(2.5)

(We assume $T_0 \ll T_b$ in this section.)

The relations on the combustion limit (2.1) follow from (2.5) and the stationary connection between the combustion velocity and the temperature in the reaction zone of form (1.10). The introduction of the parameter β permits rewriting the first relationship in (2.1) as

$$\beta^* = \frac{T_a}{T_b^*} = 1 + \frac{1}{2k} \qquad \left(k = \frac{E}{2R\Gamma_a}\right)$$
(2.6)

Let us investigate the one-dimensional (K = 0) instability by the scheme used in the previous section. The perturbations of the combustion surface and the temperatures in zones 1 and 2 are

$$\xi = D \exp \omega t$$

$$T_{1}' = A \exp \left(z_{1}' u_{b} x / 2 \varkappa \right), \quad z_{1}' = 1 + \sqrt{\beta^{2} + 4\Omega}$$

$$T_{2}' = B \exp \left(z_{2}' u_{b} x / 2 \varkappa \right), \quad z_{2}' = 1 - \sqrt{\beta^{2} + 4\Omega}$$

$$\left(\Omega = \frac{\omega \varkappa}{u_{b}^{2}} \right)$$

$$(2.7)$$

The first two relations in (1.12) remain the same; in place of the third we have

$$\frac{\partial\xi}{\partial t} = -k \frac{\beta^2 u_b}{T_a} \Big(T_2' + \frac{d T_2^{\circ}}{dx} \xi \Big)$$
(2.8)

Substituting the solutions (2.3), (2.7) into the first two relations in (1.12) and in (2.8), and taking account of (2.5), we obtain a homogeneous system of equations in A, B, and D, from which the characteristic equation for Ω follows

$$4\Omega^2 + \beta\Omega\left(\beta + 4k - k^2\beta^3\right) + k\beta^2\left(\beta - k\beta^2 + k\right) = 0$$
(2.9)

For $\beta = 1$ the relation (2.9) agrees with (1.13) if s = 0 is inserted in the latter. The equation for the stability boundary $k(\beta')$ resulting from (2.9) is

$$k = \frac{1}{\beta^{\prime *}} (2 + \sqrt{4 + \beta^{\prime *}}), \quad \beta' = \frac{T_a}{T_b'}$$
(2.10)

where T_b' is the temperature in the combustion zone corresponding to the limit of one-dimensional stability.

The dependence $\beta'(k)$ determined from (2.10) and $\beta^*(k)$ from (2.6) are represented in Fig. 2. The instability domain is located above the curve $\beta'(k)$; the domain under the curve $\beta^*(k)$ corresponds to the collapse of combustion. For this compound (k fixed), as the heat emission increases, the stationary combustion mode becomes unstable earlier than the collapse of combustion sets in because of heat transfer to the wall. This holds for 2.7 < k < 4.24 (for k > 4.24 the combustion front is unstable to one-dimensional perturbations even under adiabatic conditions). Therefore, the temperature T_b' in the reaction zone (the combustion velocity u_b' , respectively) corresponding to the loss of stability, can considerably exceed the combustion temperature at the limit of combustion T_b^* (the combustion velocity at the limit u_b^* , respectively, which is \sqrt{e} -fold less than the adiabatic value).

Constructed in Fig. 3 as a function of the parameter k is the magnitude of the combustion velocity on the stability boundary

$$u_{b}'/u_{a} = \exp\left(k\left[1 - \beta'(k)\right]\right)$$
(2.11)

3. Stability of the Flameless Combustion Model

Now, let us consider the other simplest model of combustion of a condensed material when gaseous products are formed because of reaction and the chemical transformation occurs on a solid phase surface. Such a mode, called flameless combustion, is observed in tests in the combustion of ballistic powders under conditions of very low pressure [9].

Let us analyze the stability of such a model relative to three-dimensional perturbations. The stationary temperature distributions in zones 1 and 2 are

$$T_1^{\circ} = T_0 + (T_a - T_0) \exp(u_1 x / \varkappa_1), \ T_2^{\circ} = T_a$$
(3.1)

Because of the continuity equation we have

$$\rho_1 u_1 = \rho_2 u_2 = j, \ u_1 / u_2 = \rho_2 / \rho_1 \ll 1$$
(3.2)

The equation of thermal energy balance on the powder surface

$$x = 0, \quad -\lambda_1 \frac{dT_1}{dx} + \rho_1 u_1 (q + c_1 T_a) = \rho_2 u_2 c_2 T_a$$
(3.3)

determines the stationary temperature in the reaction zone

$$T_a = \frac{c_1}{c_2} T_0 + \frac{q}{c_2}$$
(3.4)

The perturbations of the powder surface and temperatures in zones 1 and 2 have the form (1.4), (1.5). The solution of the linearized heat-conduction equation (1.6) in zone 1 yields







$$T_{1}' = A \exp(z_{1}u_{1}x/2\kappa_{1}), \quad z_{1} = 1 + \sqrt{1 + 4\Omega + s^{2}}$$
$$\Omega = \frac{\kappa_{1}}{u_{1}^{2}} \omega, \quad s = 2 \frac{\kappa_{1}}{u_{1}} K$$
(3.5)

The heat-conduction equation in the domain of gaseous reaction products

$$\rho_2 c_2 \frac{\partial T_2}{\partial t} = \lambda_2 \left(\frac{\partial^2 T_2}{\partial x^2} + \frac{\partial^2 T_2}{\partial y^2} \right) - j_x c_2 \frac{\partial T_2}{\partial x} - j_y c_2 \frac{\partial T_2}{\partial y}$$
(3.6)

contains the components j_x and j_y of the material flux along the coordinate axes. However, since the stationary temperature distribution in zone 2 is independent of the coordinates, perturbations of the quantities j_x and j_y will not originate in the linearized equation (3.6)

$$\rho_2 c_2 \frac{\partial T_2'}{\partial t} = \lambda_2 \left(\frac{\partial^2 T_2'}{\partial x^2} + \frac{\partial^2 T_2'}{\partial y^2} \right) - j c_2 \frac{\partial T_2'}{\partial x}$$
(3.7)

Substituting the expression for T_2' into (3.7) and solving the equation obtained, we have

$$T_{2}' = B \exp(z_{2}u_{2}x/2\varkappa_{2}), \ z_{2} = 1 - \sqrt{1 + s^{2}\alpha + 4\frac{\varkappa_{2}}{u_{2}^{2}}\omega}, \quad \alpha = \left(\frac{\lambda_{2}}{\lambda_{1}}\frac{c_{1}}{c_{2}}\right)^{2}$$
(3.8)

Let us note that taking account of (3.2)

$$\frac{\omega \kappa_2}{u_2^2} = \Omega \frac{\lambda_2}{\lambda_1} \frac{c_1}{c_2} \frac{\rho_2}{\rho_1} \ll 1$$
(3.9)

This permits neglecting the last member in the radicand in (3.8). The condition (3.9) denotes the non-inertia of the processes in the gas phase.

Substitution of the solutions (3.1), (3.5), (3.8) into the first equation in (1.12), which expresses the condition of temperature continuity on the combustion surface, yields

$$\frac{u_1}{x_0} \left(T_a - T_0 \right) D + A - B = 0 \tag{3.10}$$

Introducing the coefficient of temperature response of the combustion velocity in conformity with (1.9), let us write the condition of total consumption of the reacting material in the reaction front analogous to (1.11)

$$x = \xi, \quad \frac{\partial \xi}{\partial t} = -\frac{k}{T_a - T_0} u_1 \frac{c_2}{c_1} T_2'$$
(3.11)

For a combustion law of the form (1.10)

$$k = \frac{(T_a - T_0)E}{2RT_a^2} \frac{c_1}{c_2}$$
(3.12)

Substituting the expressions for ξ and T_2 ' into (3.11) yields

$$\frac{k}{T_a - T_0} \frac{c_2}{c_1} B + \Omega D = 0 \tag{3.13}$$

The energy-balance condition on the solid phase surface is in the approximation under consideration

$$x = \xi, \quad -\lambda_1 \frac{\partial T_1}{\partial x} + \rho_1 \left(u_1 - \frac{\partial \xi}{\partial t} \right) (q + c_1 T_1) = -\lambda_2 \frac{\partial T_2}{\partial x} + \rho_1 \left(u_1 - \frac{\partial \xi}{\partial t} \right) c_2 T_2 \quad (3.14)$$

Linearizing (3.14) and substituting the expression for small admixtures results in the equation

$$\left(\frac{1}{2}z_1 - 1\right)A - \frac{c_2}{c_1}\left(k - 1 + \frac{1}{2}z_2\right)B = 0 \tag{3.15}$$

Equating the determinant of the system of linear homogeneous equations (3.10), (3.13), (3.15) to zero, we obtain the characteristic equation for Ω



$$4\Omega^{3} + \Omega^{2} \left[1 + 8kr + s^{2} - (1 + 2kr - r + r \sqrt{1 + s^{2}\alpha})^{2}\right] + \Omega 2kr (s^{2} + r + r \sqrt{1 + s^{2}\alpha}) + k^{2}r^{2}s^{2} = 0$$

$$r = \frac{c_{2}}{c_{1}} = \left(\frac{dT_{a}}{dT_{0}}\right)^{-1}$$
(3.16)

The stability boundary is determined by the equation

$$k = \frac{1}{2r} \left\{ \left[1 + R - \frac{s^{2}}{2(s^{2} + R)} \right] + \sqrt{\left[1 + R - \frac{s^{2}}{2(s^{2} + R)} \right]^{2} + s^{2}(1 - r^{2}\alpha) + 2R(1 - r)} \right\}$$

$$R = r(1 + \sqrt{1 + s^{2}\alpha})$$
(3.17)

The frequency on the stability boundary is pure imaginary ($\Omega = i\psi$) and depends on the perturbation wavelength as follows:

$$\psi^2 = \frac{1}{2}kr\,(s^2 + R) \tag{3.18}$$

where k is expressed as a function of s^2 from (3.17).

An investigation of the behavior of the neutral curve (3.17) as $s \rightarrow 0$ shows that the reaction front is less stable relative to three-dimensional than one-dimensional perturbations for the following relationship between the thermophysical constants of the initial material and the products:

$$\alpha < \frac{1}{2r^2} \frac{1 + \sqrt{1+8r}}{2 + \sqrt{1+8r}}$$
(3.19)

Constructed in Fig. 4 are the dependences (3.17) and (3.18) for r = 1 and the values $\alpha_1 = 0$ (curve 1), $\alpha_2 = 0.5$ (curve 2), $\alpha_3 = 1$ (curve 3). The neutral curves are shown dashed. The unstable combustion domains are located above these curves. The k = 3 corresponds to the one-dimensional stability boundary, which agrees with the results of the theory of one-dimensional stability of powder combustion [10] which yields the following relationship between the parameters on the stability boundary $r^{-1} = (k-1)^2/(k+1)$.

A diminution in the parameter α results in expansion of the instability domain. For $\alpha < 0.4$ the system becomes more stable relative to one-dimensional than three-dimensional perturbations. The minimal boundary value $k \approx 2.91$ is reached for $\alpha = 0$, hence $s \approx 1$. A comparison between Figs. 1 and 4 permits the conclusion to be made that noninertia of the zone of combustion products results in expansion of the instability domain.

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