Concentration and Temperature Nonuniformities of Combustible Mixtures as Reason for Pressure Waves Generation

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Abstract

One of the basic principles of classical combustion theory is the assumption that the pure combustible mixture is uniform and temperature or concentration gradients are negligible. However, one may notice such a fundamentally important process as spontaneous origin of nonhomogeneous patterns due to probabilistic velocity distribution of molecules in a completely premixed matter. In effect, the existence of concentration and temperature nonuniformities (CTN) is usual for the overwhelming majority of technical devices. Imperfect operation of injector elements, heat fluxes, etc., are reasons for continuous periodic, or stochastic generation of regions with CTN. The classical approach to this type of engineering application does not seem to work well for investigation of combustion instability. Practical devices for some conditions, temperature, geometry, fuel-air ratio, etc., exhibit onset of undesirable pressure waves and subsequent oscillatory behavior. An analysis of some theoretical models is performed to take into account CTN in the reactive system. It is shown that the approach proves to be extremely helpful for understanding 1) the onset of high-frequency rocket combustion instability; 2) the "knock" phenomenon in

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the internal combustion engine; 3) the generation of strong pressure waves in communicating vessels containing a reactive mixture; and 4) pressure-wave generation in vented explosions. The results demonstrate that CTN may play an important role in combustion phenomena.

Introduction

As the time scales of chemical and gasdynamic processes are essentially different, the motion of matter resulting from thermal expansion is often not taken into account in the thermal explosion theory (Frank-Kamenetski 1967). However, in conditions close to the self-ignition of a reactive mixture, new phenomena may come into existence because the chemical and gasdynamic time scales are of the same order of magnitude.

Let us consider the evolution of a local volume of the reactive mixture preconditioned to self-ignition. Self-ignition occurs in the first instance in spots where there is an optimal combination of temperature and reactant concentration. The rest of the volume is burnt thereafter in conformity with the laws of propagation of chemical reaction waves. Self-ignition is accompanied by the generation of pressure waves that heat up and mix the pure mixture with the combustion products. The process is described by at least two time scales: chemical ($\tau_c = L/U_c$) and gasdynamic ($\tau_g = L/U_g$), where $L$ is the typical dimension of the local volume, $U_c$ is the typical velocity of the chemical reaction front, and $U_g$ is the typical gasdynamic velocity. The scale $\tau_c$ is typical of the time taken for burnout of the volume after self-ignition, whereas $\tau_g$ is typical of the time required for the pressure to become equally distributed over the field of interest. In laminar flame theory $\tau_c >> \tau_g$; inasmuch as the normal velocity of the flame is $U_c = U_g << U_g$. The time scales of the phenomenon may be separated, and the gasdynamic effects may be set aside. In nonstationary thermal explosion theory $\tau_c << \tau_g$, because fast uniform burnout of the mixture is assumed to occur throughout the volume. The condition $\tau_c \approx \tau_g$ is applicable to the propagation of a detonation wave through a reactive mixture. The flow in the detonation wave is an example of the coupled motion of a shock wave and a chemical reaction front.

In general, to meet the condition $\tau_c \approx \tau_g$, the relationship $U_c \approx U_g$ must be insured. If this takes place in a local volume, the arrival of a weak pressure wave at a fixed location may coincide with the beginning of fast ignition of the mixture. In this case, pressure-wave
amplification may be expected. The situation is possible when the pressure wave is subject to a succession of "synchronization" acts with the moments of local mixture ignition. It may give rise to the rapid, progressive amplification of the wave. At some stage of the process the pressure wave may become strong enough to cause vigorous chemical activity in a pure mixture and the mixture will detonate. Thus, under certain conditions that insure the coupled motion of the pressure wave and the wave of chemical energy release, the spontaneous generation of strong shock waves and detonation may be expected.

Zeldovich and Kompaneets (1955) have suggested the idea of carrying into effect the combustion process with $U_\infty > U_s$. The idea is to control artificially or naturally the rate of propagation of the chemical release front. For example, an explosive gas mixture could be ignited with a sequence of electric sparks, or it could be preconditioned for successive self-ignition of neighboring layers. In this way the combustion process could be stimulated with arbitrarily high apparent burning velocity. According to Zeldovich and Kompaneets (1955), to generate the Chapman-Jouquet (CJ) detonation wave in the reactive mixture, the ignition source should be moved at the rate of a self-sustaining detonation. The pressure rise in a detonation is larger than any of the other rates of successive ignition. Hence, to set up the combustion mode with $U_\infty \approx U_s$, in the local volume, the conditions for nonsimultaneous self-ignition of the neighboring layers of the mixture should be met. (For instance, in such a manner when the layers react independently according to the laws of the adiabatic thermal explosion or branching-chain processes). If that is the case, the apparent propagation of the chemical release front that Zeldovich et al. (1980) call "spontaneous propagation" appears in the volume.

The coupling mechanism between the waves of chemical release and the pressure waves is closely connected with the gradients of the local parameters which affect the rate of a chemical reaction. It follows that fluctuations are the cause of various wave patterns at conditions close to self-ignition. The spontaneous fluctuations that arise as a consequence of possible bulk thermodynamic states of a macroscopic system and the fluctuations generated by the variations of initial and boundary conditions should be distinguished. The former may promote the spontaneous origin of chain carriers in the mixture and give an impetus to the local self-acceleration of the reaction (Zeldovich 1981). Considerable deviation from a mean value of a thermodynamic parameter may occur only in a very small
spatial domain of the system. The variation of initial and boundary conditions may also result in the raising of extended portions of the mixture with spatially nonuniform distribution of local parameters (temperature, concentration, etc.). For instance, if the initially uniform pressurized, gaseous explosive mixture is suddenly expanded by the rupture of a vessel wall, the system then undergoes the sequence of nonuniform states with proper gradients of the local parameters. In the conditions close to mixture self-ignition, the gradients may bring about a nonuniform chemical release; and as a result, a number of gasdynamic wave patterns may arise in the volume.

The spatial nonuniformities in the reactive mixture appear to be potential sources of combustion instabilities that happen sometimes in the operation of technical devices. For instance, it is known as a matter of experience that, under certain conditions of mixing the fuel and oxidizer in the liquid propellant rocket engine, the process is accompanied by powerful pressure jumps that arise abruptly during normal operations (Harrje and Reardon 1972). As a rule, a high-frequency combustion instability comes into effect. The other example is the knock phenomenon in the internal combustion engine that also seems to arise because of explosion processes (Sokolik 1934). Finally, we mention the secondary shock waves that have been detected in the reaction zone of heterogeneous detonation (Ragland et al. 1968). These waves seem to give support to the stationary propagation of detonation over long distances.

The possibility of spontaneous generation of detonation waves in a spatially nonuniform explosive medium was proved for the first time by the numerical calculations of Zeldovich et al. (1970). The effect of initial temperature distribution on the evolution of the gaseous reactive mixture was considered. Gelfand et al. (1985) studied this problem parametrically and discovered the specific features characterizing the inception and transformation of various wave patterns in the system. Barthel and Strehlow (1979) have used numerical modeling to analyze the effects that appear as a result of introducing an additive with enhanced reactivity into the fuel-air mixture. The spatial distribution of the preexponential factor in the Arrhenius type of chemical reaction law was used to calculate the gradient of self-ignition delay. Gelfand et al. (1986) have investigated the role played by nonuniform reactant distribution in the evolution of the explosive mixture preconditioned to self-ignition. In all cases under review, the calculations reveal the possibility
of spontaneous generation of shock waves, their amplification, and further transition to detonation.

The phenomena in point seem to have already been observed experimentally. Borisov et al. (1970) have observed the onset of detonation after a weak shock wave passing through the cloud containing preliminary ignited kerosene droplets. The weak shock-wave-stimulated droplets break up and cause the pure mixture to mix with the combustion products, thus creating the longitudinal gradient of ignition delay. Under certain conditions the fast successive self-ignition of hot mixture portions led to generation and amplification of a secondary shock wave and finally to onset of detonation. Lee et al. (1978) have conducted experiments on photochemical initiation of detonation in mixtures of $C_2H_2-O_2$ and $H_2-Cl_2$. To meet the condition $U_\alpha \approx U_\phi$, the flash-photolysis technique was used to create the proper distribution of active radicals in the mixture. Knystautas et al. (1979) have investigated the onset of detonation by injecting the hot combustion products into the pure $C_2H_2-O_2$ mixture through perforated plates.

In this work we analyze theoretically the types of explosion processes that may be initiated in systems with spatially nonuniform distribution of temperature and reactant concentration. Of special interest are the questions relating to "critical" dimensions of the nonuniformities and sensitivity of the phenomena to variation of initial and boundary conditions. The effects accompanying multistage ignition are discussed, and some problems of practical concern are addressed.

**Formulation**

Consider a volume of typical dimension $D$ containing a multicomponent gaseous explosive mixture. For the sake of simplicity, we single out only two components: a moderately active component A and a component B with enhanced reactivity. The component B may represent an additive to the basic fuel that promotes energy release in the system or the products of preflame autooxidation such as peroxides. We assume that at the initial instant the mixture is quiescent and the pressure is uniform throughout the volume. It is also suggested that a local spatial domain of characteristic dimension $\epsilon$ (henceforth referred to as $\epsilon$-domain) exists in which the nonuniform distribution of temperature, such as

\begin{align}
T(0,R) &= T_0 - \chi T R \quad \text{in } 0 < R \leq \epsilon \\
T(0,R) &= T_e \quad \text{in } R > \epsilon
\end{align}

(1)
or the mass fraction of component B, such as
\[ B(0,R) = B_0 (1 - \chi_b R/\epsilon) \quad \text{in } 0 < R \leq \epsilon \]
\[ B(0,R) = 0 \quad \text{in } R > \epsilon \]  
(2)
is given. Here R is the Eulerian coordinate originated in the center of the \( \epsilon \)-domain, \( \chi_b \) is the temperature gradient, \( 0 \leq \chi_b \leq 1 \) is the coefficient whose value determines the form of the distribution, and subscript 0 denotes the maximum value of a parameter at the instant \( t=0 \). To simplify the problem, the component A is assumed to be uniformly distributed throughout the volume. The evolution of the system after self-ignition is assumed to be governed by the one-dimensional equations of conservation of mass, momentum, energy, and reactant species without regard for effects associated with viscosity, heat conductivity, and mass diffusion. As a consequence, the analysis is restricted to processes that come into play well before the dissipative effects gain control over the evolution. The equations can be written in terms of the Lagrangian frame as follows:

\[ \gamma \partial U/\partial \tau + (\xi/\gamma) V^{-1} \partial \theta/\partial r = 0, \quad \partial \xi/\partial \tau = U \]
\[ \partial \phi/\partial \tau = \phi \left( \frac{r}{\xi} \right) V^{-1} \]
\[ (\gamma - 1)^{-1} \partial \phi/\partial \tau + p \partial \theta/\partial \tau = \alpha_a a \exp[\beta_a (1 - \theta^{-1})] \]
\[ + \alpha_b b \exp[\beta_a - \beta_b \theta^{-1}] \]
\[ \partial a/\partial \tau + a \exp[\beta_a (1 - \theta^{-1})] = 0 \]
\[ \partial b/\partial \tau + b \exp[\beta_a (1 - \beta_b \theta^{-1})] k_b k_a^{-1} = 0 \]  
(3)
where \( p \) is the dimensionless values of pressure, \( \phi \), specific volume; \( U \), velocity; \( \theta \), temperature; \( a \), the mass fraction of component A; \( b \), the mass fraction of component B; \( \xi \), the Eulerian coordinate; \( r \), the Lagrangian coordinate and \( \tau \), is time. The dimensionless variables are defined as:

\[ p = p/p_0, \quad \theta = T/T_0, \quad U = U/\gamma R^* T_0 \]
\[ a = A/A_0, \quad b = B/B_0, \quad \tau = tk_a \exp(-E/RT_0) \]
\[ \alpha_a = Q_a/R^* T_0, \quad \alpha_b = Q_b/R^* T_0, \quad \beta_a = E_a/R^* T_0 \]
\[ \beta_b = \frac{E_b}{R_b T_0}, \xi = \frac{[Rk_a \exp(-E_a/R_b T_0)]}{\sqrt{\gamma R_b T_0}} \]
\[ r = (V \int_0^\xi \sigma^{-1} \xi V^{-1} d\xi)^{1/4} \]

where \( R_b \) is the gas constant for the mixture; \( Q \) is the chemical energy; \( E \) is the activation energy; \( k \) is the preexponential factor; \( \gamma \) is the (constant) ratio of specific heats; indices \( a \) and \( b \) denote the values related to the proper components; \( V=1,2,3 \), for plane, cylindrical, and spherical symmetry. The dimensionless temperature gradient is given by

\[ \lambda_T = \frac{\chi_T \sqrt{\gamma R_b T_0}}{[T_0 k_a \exp(-E_a/R_b T_0)]} \]

In the center of symmetry we suppose \( U=0 \) for \( \tau \geq 0 \). If the reactive mixture is placed in a vessel, it will be necessary to set boundary conditions at the walls that are assumed to be impermeable. The initial conditions \((\tau=0)\) are

\[ p = 1, \theta = 1 - \lambda_T \xi, \theta = \theta, U = 0, a = 1 \]
\[ b = 1 - \chi_b \xi / \xi_0 \text{ for } \xi \leq \xi_0 \]
\[ p = 1, \theta = 1 - \lambda_T \xi_0, \theta = \theta, U = 0, a = 1 \]
\[ b = 0 \text{ for } \xi > \xi_0 \]

(4)

where \( \xi_0 = \xi(\epsilon) \).

The system of Eq. 3 is solved numerically, subject to the initial and boundary conditions, through the use of the artificial viscosity technique (Richtmyer and Morton 1967).

Results

Four Types of Explosion Processes

To study wave patterns generated by local explosion events in the spatially nonuniform reactive medium, we consider the problem of the evolution of a two-component mixture with nonuniform initial distribution of a component B [see Eq. 2]. At \( \tau = 0 \), the temperature is assumed to be constant throughout the volume, i.e., \( T=T_0=0 \) and \( \gamma_T=0 \) in Eq. (1). Heat release resulting from the self-ignition of component B produces temperature gradients in the \( \xi \)-domain and creates conditions for the possible speeding up of pressure waves generated by the subsequent explosion of component A (Zeldovich et al. 1970). Depending on the
dimension of $\varepsilon$-domain, the kinetic parameters, and the reaction heats, four various types of explosion processes were revealed in volume D by numerical computations. Our computations were terminated when the elapsed time was as large as the typical values of diffusion or conduction time scales. The numerical values for the parameters in Eqs. 3 and 4 have been chosen as typical for combustion chambers:

$$\gamma = 1.2, \ 800 \leq T_0 \leq 1400^\circ K,$$

$$k_a = k_b = 10^{10} s^{-1}, \ 5 \leq \alpha_a \leq 8,$$

$$3 \leq \alpha_b \leq 5, \ 15 \leq \beta_a \leq 20,$$

$$12 \leq \beta_b \leq 15, \text{ and } 0.2 \leq \epsilon \leq 100 \text{mm}.$$

The four types of explosion processes that may be generated spontaneously in the system are described in the following discussion.

**First Type.** If $\epsilon \rightarrow D$ and $\chi_b \rightarrow 0$, then the constant volume explosion takes place in the system with almost uniform pressure rise throughout the volume. For the given numerical values of reaction heats, the pressure has been increased by no more than a factor of 3, compared with the initial pressure.

**Second Type.** If $\alpha_* < \epsilon < D$ and $\chi_b = 1$, the range of basic parameters exists in which the onset of detonation inside the $\varepsilon$-domain occurs and further stationary propagation of the detonation wave takes place in the initially uniform medium. The temporal evolution of such a flow is presented in Fig. 1, which shows pressure $P$ against distance $\xi$ for several values of $\tau$. The mixture is defined

![Fig. 1](image)

The onset of detonation in the system with distributed additive B. Shock pressure-time history in dimensionless time interval $\Delta t = 0.27$. 

by the following values of the basic parameters: $\beta_a = 16.7$, $\beta_b = 12.5$, $\alpha_a = 8$, $\alpha_b = 3$, $T_0 = 1000$°K, and $\epsilon_0 = 50$ mm. The features of the evolution are the same as described by Zeldovich et al. (1970). The pressure rise behind the detonation wave is more than eight times greater that the initial pressure. This type of explosion process is important for the following analysis.

**Third Type.** If $\epsilon_{**} < \epsilon < \epsilon_*$, then in a certain range of basic parameters the detonation wave was generated outside the $\epsilon$-domain with further stationary propagation in uniform mixture. The temporal evolution of the process may be broken into the following main points: a) self-ignition of component B and fast subsequent ignition of component A contained in the $\epsilon$-domain; b) decay of the precursor pressure wave generated by the local explosion; c) a secondary explosion of component A and formation of a secondary shock wave-reaction front complex behind the decaying precursor shock wave; and d) a secondary shock-wave reaction-front complex, which overtakes the precursor shock wave, triggering detonation.

The third type of explosion process is notable for the precursor shock wave that is precipitated by the microexplosion in the $\epsilon$-domain. The precursor wave creates the conditions for secondary wave amplification resulting from a time-phased local energy release. Figure 2 shows the temporal evolution of the mixture defined by the following values of the basic parameters: $\beta_a = 16.7$, $\beta_b = 12.5$, $\alpha_a = 8$, $\alpha_b = 3$, $T_0 = 1200$°K, and $\epsilon = 5$ mm.

![Fig. 2](image)

*Fig. 2* The onset of detonation in the system with small spatial nonuniformity of reactant concentration ($\Delta r = 0.27$). Curves 1 and 2 are drawn for $\tau = 0.326$ and $\tau = 2.49$, respectively.
Figure 3 displays the history of peak pressure in the volume. We point to the fact that the smaller the \( \epsilon < \epsilon_* \) (other things being equal), the larger the distance from the origin required for the onset of detonation. The oscillations of \( P_{\text{max}} \) arise in the numerical study as a consequence of the longitudinal instability of a one-dimensional detonation wave. The temporal shift at the instant detonation occurs is explained by the decay of the precursor shock wave and hence by the diminishing overall temperature level behind it. For \( \epsilon = \epsilon_** \approx 2 \text{ mm} \) and \( D = 100 \text{ mm} \), the detonation wave appeared on the rim of the vessel wall. For \( \epsilon < \epsilon_** \), a nonstationary explosion wave propagates throughout the volume. The pressure rise in such a wave is greater than for the case of the constant volume explosion. For \( \epsilon < \epsilon_** \), the explosion processes of the first type have been observed again.

Fourth Type. This type of explosion process is termed the oscillatory one. The computations revealed that in a certain range of basic parameters, the wave pattern shown in Fig. 4 was realized. The figure is drawn for the mixture defined as follows: \( \beta_a = 19.8 \), \( \beta_b = 14.8 \), \( \alpha_a = 7.9 \), \( \alpha_b = 2.96 \), \( T_0 = 1010^\circ \text{K} \), and \( \epsilon = 50 \text{ mm} \). The specific feature of such processes is the existence of the decaying phase in the history of a shock wave that initially gained momentum. Figure 4 represents the case when the detonation wave is initiated in close proximity to \( \epsilon \)-domain boundary, is switched off immediately, and passes to the outer region because of relatively low temperature of the region. The decaying shock wave serves as a precursor (see 3rd type)
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Fig. 4 Oscillatory explosion process. SE, second explosion. The time interval is $\Delta t_0=0.027$.

Fig. 5 The temporal evolution of pressure for oscillatory explosion processes. Curve 1 is representative of "galloping" detonation ($T_0=1013\,\text{K}$, $\beta_a=19.75$, $\beta_b=14.8$, $\alpha_3=7.9$, and $\alpha_4=2.96$). Curve 2 is indicative of detonation fail ($T_0=1012\,\text{K}$, $\beta_a=19.8$, $\beta_b=14.9$, $\alpha=7.9$, and $\alpha_b=2.96$). Curve 3 is representative of shock-wave decay in chemically inert medium.

and creates the conditions for amplification of a secondary shock wave.

Figure 5 shows the typical plots of peak pressure in the system against dimensionless time for this type of explosion process. Curve 1 corresponds to the "galloping" detonation that arises in a very narrow range of basic parameters. Dashed line 3 reflects the history of shock-wave decay in a chemically inert medium. It can be seen
from Fig. 5 that strong shock waves are periodically being generated and decaying in the volume. The interval between subsequent secondary explosions is governed by ε and kinetic parameters of the mixture. Depending on the initial condition, the detonation wave or a series of shock waves reaches the wall.

Of great importance is the sensitivity that the explosion processes under investigation have to initial temperature variations. A temperature increase from $T_0=1010^\circ\text{K}$ to $T_0=1020^\circ\text{K}$ for the mixture whose evolution is presented in Fig. 4 led to the onset of stationary detonation like that shown in Fig. 1. For temperature near $T_0=1015^\circ\text{K}$, the wave pattern was strongly dependent on $T_0$ and even on parameters of the numerical scheme (e.g., the coefficient of artificial viscosity). However, this does not mean that the chosen finite difference scheme is inadequate. There is evidence that in proximity to $T_0\approx1015^\circ\text{K}$, the solution with the second type of explosion process becomes globally unstable.

Thus, our calculations show that spontaneous generation of shock and detonation waves may occur in spatially nonuniform reactive media. Fuel droplets in heterogeneous mixtures may have an effect on the propagation of an explosion wave inasmuch as temperature and vapor mass fraction gradients are, as a rule, inherent in the neighborhood of them. However, the small dimensions of gas-phase nonuniformities required for initiation of rather strong shock waves are indicative of a mechanism capable of providing some insights about the onset of nonlinear disturbances in both homogeneous and heterogeneous reactive systems.

The other question is the critical dimension of local nonuniformity for a given system. Although our attention has been restricted to isolated local spatial nonuniformity of temperature and reactant concentration, calculations were made with provision for two or more such local nonuniformities in the one-dimensional geometry. The situation is particularly interesting when the portions of additives, each taken separately, do not promote spontaneous generation of strong pressure waves. Our study shows that, under certain conditions and at the proper distance between the centers of local nonuniformities, the strong shock and detonation waves may arise spontaneously, proceeding from the hot spots formed by collisions of the precursor shock waves. These trends were also observed when the local spatial nonuniformities of temperature and reactant concentration were displayed near the wall or a contact surface of different gases. Thus, we conclude that
the presence of distributed spatial nonuniformities of chemical activity, contact surfaces, and boundaries makes for easier initiation of strong nonlinear pressure disturbances in explosive media.

**Initial Temperature Gradient**

The case of nonuniform initial temperature in the volume while species gradients are absent [i.e., $B_0=0$ in Eq. (2)] has been considered by Zeldovich et al. (1970) and Gelfand et al. (1984). In the following discussion the criterion for the spontaneous generation of strong pressure waves is derived. Self-ignition of the explosive mixture is assumed to occur well before the spreading or deformation of the temperature profile because of heat conduction and convection. The condition $U_c=U_g$ can be written in the form

$$\frac{dR}{dt_i} \approx \sqrt{\gamma R_* T_e}$$  \hspace{1cm} (5)

where $t_i$ is the local induction time of the mixture. The derivative $dR/dt_i$ defines the velocity of the spontaneous propagation of the chemical release front. The term $dR/dt_i$ can be evaluated assuming that $t_i \approx t_{ad}$, where $t_{ad}$ is the adiabatic ignition delay. The use of the well-known expression for $t_{ad}$ (Frank-Kamenetski 1967) leads to

$$\frac{dR}{dt_i} \approx \left[ \frac{\gamma-1}{\gamma} \right] \left[ \frac{Qk}{c_i R_*} \right] \exp(-E/RT)$$  \hspace{1cm} (6)

![Fig. 6](image-url)

*Fig. 6* Calculated velocity of peak pressure point against distance to the origin of temperature nonuniformity redrawn from [Gelfand et al. (1985)]. Vertical bars on the curves give the position predicted by Eq. (7).
Equations 5 and 6 together show that the chemical and gasdynamic time scales will be the temperature given by

$$T_c = \frac{(E/R_\sigma)}{\ln[(\gamma-1)\rho k/\gamma R_\sigma \sqrt{\gamma T_c T_0}]}$$  \hspace{1cm} (7)

Amplification of a pressure wave generated by local explosion is possible in the range $T_c < T_c < T_0$. If $T_c < T_0$, then the wave patterns are very weak and almost uniform burnout of the mixture takes place throughout the volume. For temperatures approaching $T_c$ decoupling of the motion of pressure waves and chemical release front may be expected. Hence, the critical value of temperature gradient $x_t = x_t^*$ exists such that, for $x_t > x_t^*$, the phase of pressure-wave amplification that results from interlocking with the local ignition event is absent. Thus, we conclude that the generation of strong pressure waves may be expected in the range $T_e < T_c < T_0$ for $x_t < x_t^*$. For $x_t > x_t^*$, within the limits $T_e < T_c < T_0$ as well as for $T_c > T_0$ the local explosion with further flame front formation may be expected.

Figure 6 taken from Gelfand et al. (1986) shows the dependence of the peak pressure point velocity $U_m$ in the flow from distance to the origin. The numerical values that have been used for the parameters in the problem are $x_t = 2.5 \times 10^{-6}$ K/m, $T_0 = 2000$ K, $K = 10^{10}$ s$^{-1}$, $V = 1$, $\gamma = 1.2$, and $E/R_\sigma = 10$.

The vertical bars on the curves give the positions $R = R_c$ corresponding to the values of $T_c$ that have been calculated on the basis of Eq. 7. The lower curve represents the case $x_t = x_t^* = 0$. Thus, it stands to reason that Eq. 5 reflects the main features of the phenomena that may occur in conditions close to self-ignition.

Influence of Additive

The effects produced by an additive with enhanced reactivity placed into $\epsilon$-domain in a stepwise distribution are now considered. Thus, we assume that $x_h = x_h^* = 0$ in Eqs. 1 and 2 and that $T_a = T_0$. We will show that in this situation not only explosion processes of the first and third types may make an appearance, but also processes of the second type may arise with the onset of detonation inside the $\epsilon$-domain. The analysis under development may prove to be useful for understanding the mechanism of knock phenomenon in the internal combustion engine and for clearing up the possible combustion modes that may occur inside fuel-air clouds. In this instance the rarefaction wave propagates through the $\epsilon$-domain after explosion-like self-ignition of component B, producing the conditions for rapid
amplification of the pressure wave generated by subsequent explosion of component A. The analysis is based on Eq. 5. Fast self-ignition of component B serves to increase the temperature to \( T_b \approx T_e + (\gamma - 1)Q_b/R_\star \). For the first stage of ignition process \( \alpha_b \approx 1.0 \); i.e., the reaction heat is relatively small. This is especially true for both the additives promoting combustion and the cold-flame stage of low-temperature self-ignition of complex explosive mixtures.

As a result of thermal expansion of \( \epsilon \)-domain, a weak pressure wave is generated in the outer region and a rarefaction wave propagates inside the domain. Temperature reduction at the contact surface may be easily evaluated from the elementary theory of shock tubes (Gaydon and Hurle 1963). With the acoustic approximation gives the temperature reduction is

\[
\Delta T \approx \frac{\gamma - 1}{\gamma} T_b \left( \sqrt{\frac{T_b}{T_e}} - 1 \right)
\]

The typical temperature gradient formed in the system before self-ignition of component A may be written as

\[ x_T \approx \frac{\Delta T}{\epsilon} \quad (8) \]

To obtain the condition of pressure-wave amplification after subsequent self-ignition of component A, \( x_T \) [Eq. 8] is equated to the temperature gradient given by Eqs. 5 and 6:

\[
(T_b/\epsilon) \left( \sqrt{\frac{T_b}{T_e}} - 1 \right) = \left( \frac{Q_a}{R_\star} \right) \left[ k_a \exp \left( -\frac{E_a}{R_\star T_b} \right) \right]/\sqrt{\gamma R_\star T_b}
\]

After transforming and taking natural logarithms of both parts of Eq. 9 and neglecting small terms, the following condition of pressure-wave amplification can be obtained:

\[ T_b \approx \frac{E_a}{R_\star \ln (Q_a k_a \epsilon/\sqrt{\gamma R T_e})} \]

or, when rewritten in dimensionless quantities:

\[ \theta_b \approx \frac{\beta_a}{\ln (\alpha_a k_a \epsilon/\sqrt{\gamma R T_e})} \quad (10) \]

On the one hand, Eq. 10 allows the evaluation of the minimum spatial dimension \( \epsilon = \epsilon_C \) of an additive portion with the enhanced reactivity required for triggering of the strong shock waves. On the other hand, Eq. 10 allows the determination of the heat of a subsidiary reaction \( Q_b \).
whose value is required to switch on the phenomenon

$$\alpha_b = (\delta - 1)/(\gamma - 1)$$

where

$$\delta = \frac{\beta_a}{\ln(\alpha_a k_a e^{1/2} / \sqrt{\gamma R T_e})}$$

If the additive is absent (i.e., $\alpha_b = 0$) and the mass fraction of component A is initially zero outside the $\epsilon$-domain, then Eq. 10 allows the evaluation of initial temperature $T_e$, for which the explosion type of self-ignition of component A will be accompanied by amplification of the generated shock wave. This temperature may be given by the following transcendental equation:

$$1 + \beta_a^{-1} \approx \delta$$

where the value of typical preignition heating $R_e T_e^{2/3}$ is used for initial heating of the mixture. In Fig. 7, Eq. 11 is represented by a dashed line. Our numerical

![Graph](image-url)
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calculations show that in the coordinates plotted along the axes of Fig. 7 the coupled motion of a pressure wave and a chemical release front occurs only in the dashed segment. As a matter of convenience it is assumed in calculations that the mixture outside the ε-domain is chemically inert. Near the lower boundary line of the segment, a shock wave lifts pressure to a maximum value at the contact surface that now may achieve the value of peak pressure in the CJ detonation wave. The upper boundary line of the segment is somewhat conditional, since it is drawn to meet condition

\[ P_{\text{max}} = 1.1[1 + (\gamma - 1)(\alpha_a + \alpha_b)]. \]

It is evident from Fig. 7 that the lower boundary line of the segment does not begin at the origin of the coordinates. The boundary lines of the segment displace only slightly under a variation of the type of symmetry.

Expansion

An analysis of a Lagrangian problem, i.e., the problem of accelerating a body by expanding combustion products is presented in the following. The main objective is to elucidate the role played by a rarefaction wave appearing in the wake of a moving body. The reactive mixture, which is confined in the volume \( V_0 = S\epsilon \) (where \( S \) is the cross-sectional area, and \( \epsilon \) is the longitudinal dimension of a channel) between the impermeable wall from the left and a piston of mass \( M \) from the right, is assumed to undergo a one-step, first-order Arrhenius type of chemical reaction as written for component A in Eq. 3. At the initial instant the state of the mixture is uniform [i.e., \( \chi_A = \chi_B = 0 \) in Eq. 4] and is defined by the values of pressure \( P_0 \) and temperature \( T_0 \), whereas at the piston surface there is a vacuum, i.e., \( P = 0 \) at the piston. For the sake of simplicity friction and heat losses are neglected when the piston is moved along the semi-infinite channel. The quantities of interest are piston velocity, maximum pressure rise, and impulse acting on the wall being associated with the motion of both the mixture and the piston.

To solve the problem, we use Eq. 3, with all terms involving the component B and the equation for component B omitted. The position of a closed-end of the channel is at \( R = 0 \). The dimensionless mass of the piston is defined as

\[ \mu = \frac{M}{\gamma R \frac{S P_0}{T_0}} [k \exp(-\beta)]/\gamma P_0 S \]
and write down the boundary conditions at the piston:
\[ \xi = \xi_0: \frac{dU}{d\tau} = p/\gamma \mu \]

Thus, the problem is formulated through the use of Eqs. 3 and 13 and initial conditions
\[ \tau = 0, \xi < \xi_0: p = \theta = a = 1, U = 0 \]

The motion of the piston causes a rarefaction wave to propagate in the mixture. If inert gas is considered, the well-known expressions for a rarefaction wave in the time interval \(0 < \tau < \xi_0\) are
\[ \xi - \omega = (U - \theta^{0.5})(\tau + \omega) \]
\[ U = 2(1 - \theta^{0.5})/(\gamma - 1) \]

where \(\omega = 2\gamma \mu/(\gamma + 1)\). The temperature distribution in an inert rarefaction wave is:
\[ \theta = (\frac{2}{(\gamma - 1)}(\xi - \omega)/(\tau + \omega))/((\gamma + 1)) \]

Let \(\tau_{ad} = \gamma/(\gamma - 1)\alpha \beta\) be the adiabatic ignition delay of the reactive mixture at the initial temperature \(\theta = 1\). Then, for \(\tau < \tau_{ad}\) and \(\beta >> 1\), the distribution given by Eq. 16 is valid for a rarefaction wave in an explosive medium.

If \(\tau_{ad} < \xi_0\), then at the instant \(\tau = \tau_{ad}\), a constant volume explosion takes place in the interval \((0, \xi_0 - \tau_{ad})\). For \(\beta >> 1\) the overpressure produced by such an explosion is \(\Delta p = (\gamma - 1)\alpha\). The pressure jump that thus appears serves to form a compression wave in the system.

The temperature distribution of Eq. 16 results in a gradient of ignition delay increasing toward the piston and brings about nonuniform chemical energy release along the length of the channel. The process of successive self-ignition of portions of the mixture may be considered as propagation of a chemical release front.

The variation of gas temperature at a piston surface for the instant of time \(\tau = \tau_{ad}\) is given by the expression
\[ \Delta \theta_p = 1 - (1 - (\gamma - 1)/2)U_p)^2 \]

where, the subscript \(p\) denotes the quantities at the piston surface and \(U_p\) is the piston velocity. If \(\Delta \theta_p\) is equal to \(\beta^{-1}\), the typical scale of temperature difference for explosion processes, the conditions close to the self-ignition limit are fulfilled in the neighborhood of the
piston. Hence a distinction between the two following limits arising in the problem can be made.

The first limit is $\Delta \theta_p << \beta^{-1}$. In this case the velocity of the chemical release front is much greater than the typical gasdynamic velocity, and a constant volume explosion consumes the mixture with almost uniform pressure rise along the length of the channel. If we take into account that $U_p \approx \tau_{ad}/\gamma \mu$, the condition under examination may be written in the form

$$1 - \left(1 - \frac{(\gamma - 1)/2}{\tau_{ad}/\gamma \mu}\right)^2 << \beta^{-1}$$

As long as $\beta^{-1} << 1, \alpha \mu >> 1$. The same condition may be derived from the exact expression for $U_p$ in the rarefaction wave.

Self-ignition may also occur for $\tau_{ad} > \xi_0$. At the instant $\tau = \xi_0$, the head of the rarefaction wave is reflected from the closed end of the channel. The maximum decrease in temperature in the reflected rarefaction wave is achieved at the time $\tau \approx \tau_1 = 2\xi_0 + \xi_0^2/\omega$ and is given by Eq. 17. Here, $\tau_1$ is the time taken for the reflected wave to reach the piston in chemically inert gas. If ignition occurs at that time, Eq. 18 remains valid.

For $\tau_{ad} > \tau_1$, the wave reflected from the piston should be taken into account. Inasmuch as we consider small decreases in temperature for the time of two reflections, we may assume the stationary temperature variation in the channel. In this case the condition $\theta_p << \beta^{-1}$ will be given by

$$1 - \left(1 + \Delta \xi/\xi_0\right)^{-1} (\gamma - 1) << \beta^{-1}$$

Since $\Delta \xi \approx \rho_p \tau_{ad}/\gamma$, $\alpha \mu >> \tau_{ad}/2\xi_0 > 1$ from Eq. (18).

Thus, the expression $\Delta \theta_p << \beta^{-1}$ is equivalent to Eq. (18). For the case in point, the value of the maximum pressure is less than or equal to $P_{max} = 1 + \Delta P$. In contrast to the Lagrangian problem with momentary energy release, the maximum piston acceleration in this instance is achieved at $\tau \geq \tau_{ad}$. Hence, the impulse acting on channel walls changes stepwise after the reaction front reaches the piston.

It should be noted that Eq. 18 contains some typical mass $\rho_0 S/\rho_R T_0/[k \exp(-\Theta)]$ instead of the total mass of the mixture. Here $\rho_0$ is the initial density of the gas. This quantity corresponds to the mass of the mixture involved in motion by time $\tau = 1$, which is equal to the chemical time scale.
The second limit is $\alpha \mu \ll 1$. In this case, velocity of
the heat release front at $\tau \geq \tau_{ad}$ is much less than the
typical gasdynamic velocity. Indeed, for $\beta \gg 1$ we have

$$\alpha \mu = \gamma \mu / (\gamma - 1) \tau_{ad} \beta$$

$$\approx [(\partial \xi / \partial \theta)_{\tau=\tau_{ad}} / (\partial \xi / \partial \theta)_{\theta=0}] = \partial \xi / \partial \tau_{i} \ll 1$$

$$\xi = -\tau_{ad}$$

where $\tau_{i} = \tau_{ad} \theta^{2} \exp[-\beta(1-\theta^{-1})]$ is the induction time at
temperature $\theta$. The derivative $\partial \xi / \partial \tau_{i}$ determines a
velocity of the heat release front as well as the typical
gasdynamic velocity, where $\theta \ll 1$.

In the case under study the maximum pressure rise is
also not greater than $P_{\text{max}} = 1+\Delta P$ for all the time the
piston is accelerated. After the self-ignition event, a
decaying shock wave propagates through the mixture. The
arrival of the shock wave at the piston surface leads to a
change in its motion and a varying impulse acting on the
channel walls. Shock-wave loading of the piston may
generate inertial effects with piston velocity jumps.

In the intermediate case ($\alpha \mu \omega l$) the amplification of
a shock wave due to coupling with a local chemical release
may be expected after self-ignition. In accordance with
the results of section 1, the onset of detonation may occur
in this case, and secondary explosions and decaying
precursor shock waves may also occur. Reflection of the
detonation wave from the piston may lift the pressure level
to such an extent that the channel walls may burst. The
additional effect on the channel walls is connected with
longitudinal impulsive loading caused by the periodically
accelerating motion of a body after the stroke.

Next, we will derive the condition for cessation of
self-ignition. The critical condition of self-ignition at
$\tau = \xi$, may be obtained by equating the rate of heat release
in the mixture with the rate of cooling the mixture in the
rarefaction wave. In keeping with Eq. 16 the rate of
cooling in the first rarefaction wave is given by ($\tau = \xi_{0}$,
$\xi = -\xi_{0}$)

$$\theta = -2(\gamma - 1) / [(\gamma + 1)(\xi_{0} + \omega)]$$

The rate of temperature rise due to chemical energy release is

$$\partial \theta / \partial \tau = \exp[\beta(\theta - 1)]$$

$$\beta \tau_{ad}$$
Since $\beta(\theta-1) = -\ln(1-\xi_0/\tau_{ad})$, we have

$$\partial \theta/\partial \tau = [\beta(\tau_{ad} - \xi_0)]^{-1}$$

The critical condition may then be written in the form

$$2(\gamma-1)/[(\gamma+1)(\xi_0+\omega)] = \beta(\tau_{ad} - \xi_0)^{-1}$$

or after transformation,

$$1-\alpha \mu \geq (\xi_0/\tau_{ad})[1 + (\gamma+1)/2\beta(\gamma-1)]$$

Equation 19 is true for the first rarefaction wave and is also valid for a reflected wave ($\tau > \xi_0$). As evidenced by Eq. 19, self-ignition is possible when two conditions are met:

$$\alpha \mu << 1$$

and

$$\tau_{ad} \leq \xi_0[1 + (\gamma+1)/2\beta(\gamma-1)]$$

For $\alpha \mu >> 1$ and $\beta >> 1$, diminution of temperature in a rarefaction wave is $\Delta \theta \approx \beta^{-1}$, and self-ignition always occurs.

Thus, we may give the four possible models of system evolution distinguished by a value of the parameter $\alpha \mu$:

1) For $\alpha \mu << 1$ and $\tau_{ad} > \xi[1 + (\gamma+1)/2\beta(\gamma-1)]$, self-ignition does not occur, and motion of the piston is that of the case of inert gas expansion.

2) For $\alpha \mu << 1$ and $\tau_{ad} \leq \xi[1 + (\gamma+1)/2\beta(\gamma-1)]$, a decaying shock wave propagates all along the mixture.

3) For $\alpha \mu = 1$, a shock wave is generated as a result of self-ignition, which accelerates because of coupling with local energy release.

4) For $\alpha \mu >> 1$, almost uniform burnout of the mixture takes place, and shock waves are absent.

Figure 8 shows the four parametric domains. The plot is drawn on the basis of numerical solution of Eq. 3 with the initial and boundary conditions [Eqs. 13 and 14]. Along the $y$ axis the ratio of the typical gasdynamic time scale to the typical chemical heating time scale $\Pi = \xi_0/\tau_{ad}$ is plotted. Along the $x$ axis the ratio of the total mass of the gas to the piston mass $m/M = P_0 V_0/MRT_0$ is plotted.

The boundary line separating domains 1 and 2 fits well with the equation

$$\Pi = mM^{-1}/[(\gamma/\beta(\gamma-1)) + mM^{-1}[1 + (\gamma+1)/2\beta(\gamma-1)]]$$

which can be obtained from the critical condition, Eq. 19. Domain 2, which is representative of the parametric region
with decaying shock waves, is narrow compared with the other domains. Such flow patterns may arise under large gradients of induction time or in conditions close to the limit of self-ignition. The boundary line separating domains 3 and 4 is conditional to some extent. In the case under consideration the boundary line is drawn through the points in which the maximum pressure is not larger than \( P_{\text{max}} = 1.1(1+\Delta p) \).

The phenomena typical for domain 3 are very sensitive to small variations of temperature and slight additives that affect the rate of chemical release. It is evident from Fig. 8 that solutions fall into this domain only for a narrow range of \( T_0 \). For instance, for \( m/M=0.1 \) and \( \beta=20 \), and \( T_0=1000 \text{ K} \), variation of the initial temperature by 50 K leads to cessation of an amplification phase in the evolution of a compression wave and the solution falls into domain 1 or 4.

The calculated time interval required for amplification of the compression wave depends on the quantities \( \Pi \) and \( m/M \). For \( \Pi=1 \) and \( m/M=1 \) it is about a hundredth of a typical time scale of chemical reaction \( t_c = 1/k \exp(-\beta) \). Therefore, the processes under investigation are stable with respect to low-frequency acoustic oscillations and turbulent fluctuations whose characteristic time scales exceed the amplification time. Moreover, such oscillations and fluctuations may become a cause of shock wave generation under normal operating
conditions in a compression machine operated in parametric domain 1.

The present model is indeed in qualitative agreement with experimental observations of the combustion process in an internal combustion engine. In accordance with the work of Sokolik (1934), as the compression ratio is increasing, the knock phenomenon tends to gain momentum at first, with later attenuation and cessation. In the framework of the model in question, the knock phenomenon begins in domain 2 in Fig. 8. As temperature and pressure increase in the channel, the effect of explosion tends to increase too (see domain 3). Further increase of the compression ratio results in a pressure drop in the lead shock wave, and ultimately the phenomenon ceases (domain 4).

Figure 9 shows the calculated piston velocity history for m/M=1. Curve 1 corresponds to the case of a piston set in motion by a gaseous mixture with low reactivity (2 = 10^{-5}), during which time piston acceleration self-ignition does not occur. Curve 1 appears to be in good agreement with the analytical solution of the Lagrangian problem for the case of inert gas expansion. Curve 3 is representative
of the piston velocity history for the gaseous mixture with high reactivity (\( \Pi = 10^4 \)). As distinct from curve 1, the greatest acceleration in the instance is achieved at the moment \( \tau = \tau_{ad} > 0 \). For \( \Pi \to \infty \), the piston velocity appears to be in good agreement with the theory of acceleration of a body by chemically inert combustion products. Curve 2 is drawn for \( \Pi = 2.5 \), which insures that the characteristic point falls into domain 3 in Fig. 8. At the instant \( \tau = 0.062 \), a stepwise acceleration of the piston occurs (seen in Fig. 9). This is a result of detonation wave reflection from the moving piston. From this point on the motion of the piston becomes oscillatory because of inertial effects of the gaseous volume.

The point of particular interest is the maximum piston velocity (for \( \tau \to \infty \)), when \( m/M \) becomes constant for different flow patterns in the channel. Our calculations show that the maximum piston velocity increases as \( \Pi \) increases, and the greatest value is achieved for the momentary heat release at \( \tau = 0 \) (\( \Pi \to \infty \)).

Conclusion

The present analysis has demonstrated that the spontaneous generation of shock and detonation waves may occur in conditions close to self-ignition of the reactive mixture. A variety of factors studied may give an impetus to the onset of pressure waves. Among them are the temperature nonuniformities and/or portions of the mixture with distributed reactant concentration, boundaries and contact surfaces, and variation of initial and boundary conditions. It is obvious that the results obtained are valid not only for the initially quiescent reactive mixture but also hold true for a stream. An additional factor that may give rise to the phenomena mentioned above is the nonuniform residence time distribution of mixture portions in a combustion chamber. The results may provide insight into the onset of nonlinear combustion instability in various technical systems as well as the transition to detonation in accidental explosions.

References


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