Dynamic Response of a Plane-Symmetrical Exothermic Reaction Center

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The concept of the exothermic reaction center is based on the premise that in a chemically homogeneous combustion system the particles undergoing the bulk of heat release are confined within discrete small regions where, as a consequence of the inherent nonhomogeneity of the various physical phenomena that influence the induction process, the reaction proceeds preferentially, ahead of their immediate surroundings. In the present paper the dynamic response of plane-symmetrical centers is treated analytically. In this connection the conservation equations for the reaction center, consisting of a kernel where the chemical reaction takes place and its immediate surroundings which are then essentially inert, are combined yielding a single integral equation expressing, in effect, a nonlinear transfer function of the system for which the input is provided by a given time profile of the heat released per unit mass while the output gives the pressure pulse it generates under the restriction of planesymmetrical motion. The solution is governed by a Damköhler number $r_k^0/(a_s^0\delta_\omega)$ where r_k^0 is the initial radius of the kernel of the reaction center, a_s^0 the initial sound speed in the surroundings, and δ_ω the rise time of the power pulse. Thus, for a given form of the exothermic power pulse profile, the dynamic behavior of the system is completely specified in terms of only the above Damköhler number and the heat of reaction per unit mass of the combustible medium. Specific solutions are worked out for a set of typical elementary power pulse profiles, and the practical significance of the results is illustrated by their application to the problem of transition to detonation in an explosive gas.

Introduction

EXPERIMENTAL observations of autoignation in premixed gaseous mixtures indicate that the concept of a perfectly homogeneous ignition throughout the reacting medium is practically unrealizable. Instead, ignition sets in first, as a rule, within some discrete small regions of the combustible mixture where, as a consequence of the inherent nonhomogeneity of the various physical phenomena associated primarily with diffusion, the induction process proceeds faster than in their immediate surroundings. Moreover, as shown in Ref. 1, in an explosive gas the average duration of the bulk of heat release is at least one order of magnitude shorter than that of any significant diffusional effect that may concurrently take place in the reacting medium.

The system of the small kernel where the reaction occurs and its immediate surroundings is referred to as the reaction center. As a consequence of the sharpness of the exothermic power pulse, the bulk of the chemical reactions associated with heat release take place in the kernel with a negligible amount of mass and heat transfer with the surroundings. In the analysis of the dynamic behavior of the reaction center it is assumed, therefore, that the only manner in which the kernel can interact with its surroundings is by momentum transport, while the effects of the mass and heat transfer can be completely neglected.

The exothermic reaction center can be thus looked upon as a diagonally opposite model of an elementary constituent of the combustion system to that of a flame whose existence depends crucially on mass and heat transfer, whereas the momentum transport is usually considered to be of negligible influence. Although the two models bound the behavior of the actual system, the main virtue of the exothermic reaction center is that, in contrast to the flame, it provides an analytically tractable model for the determination of the dynamic behavior of the system.

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In our previous publications on this subject we provided rational criteria for specific definition of exothermic reaction centers[1] and demonstrated their physical significance.[2] This yielded, in particular, information on the time profiles of the power pulse of energy release corresponding to a given kinetic chain reaction mechanism and the accompanying pressure pulse. Under usual conditions, the average duration of these pulses was found to be of an order of a microsecond.

The statistical nature of the autoignition process in hydrocarbon-air mixtures was studied experimentally by Terao.[3,4] The smallest admissible volume of explosive reaction centers was evaluated, on the basis of chemical kinetic considerations, by Van Tiggelen,[5] while an investigation of the thermodynamic properties of the reaction centers, as they are affected by various chemical kinetic processes, was carried out recently by Borisov.[6]

Here the dynamic response of plane-symmetrical centers is treated analytically. In this connection the conservation equations of the exothermic reaction center are combined to yield a nonlinear transfer function for which the input is provided by a given power pulse of energy release and the output is expressed in terms of the pressure pulse it generates under the restriction of plane-symmetrical motion.

The transfer function is expressed in terms of an integral equation with a Damköhler number $r_k^0/(a_s^0\delta\omega)$, where r_k^0 is the initial radius of the kernel of the reaction center, a_s^0 is the initial sound speed in the surroundings and δ_ω is the rise time of the power pulse, acting as a similarity parameter. On this basis it is demonstrated that, for a given form of the time profile of the exothermic power pulse, the dynamic behavior of the system is completely specified in terms of only the above Damköhler number and the heat of reaction per unit mass of the combustible medium.

Specific solutions are then given for a set of typical elementary time profiles of heat release, or of the exothermic power pulse. The practical significance of the results thus obtained is illustrated by their application to the problem of transition to detonation in an explosive gas mixture.

System

The analysis presented here of the exothermic reaction center is based on the following assumptions: 1) The reacting system is considered to consist of two regions: the kernel where the exo-

thermic reaction occurs, and the surroundings which are then essentially inert, being subjected only to an adiabatic compression due to the expansion of the kernel. 2) The interface between the kernel and the surroundings is impervious to all transport phenomena except for momentum transfer. 3) All the processes within the kernel occur in bulk so that at every instant of time its properties are spatially uniform. 4) The flow system is plane-symmetric so that the compression of the surroundings is performed by a simple wave. 5) Substances in both regions are treated as perfect gases with constant specific heats. 6) Prior to the start of the reaction, the medium throughout the center is homogeneous and quiescent. Under such circumstances, the dynamic behavior of the system is controlled by the continuity and energy equations for the kernel, and the characteristic equation for the simple wave in the surroundings. These can be expressed, respectively, as follows:

$$r_k = (v_k/v_k^0)r_k^0 \tag{1}$$

$$[1/(\gamma_k - 1)]\, d(p_k v_k)/dt + p_k\, dv_k/dt = dg/dt \tag{2}$$

where

$$u_s = [2a_s^0/(\gamma_s - 1)][(p_s/p_s^0)^\alpha - 1] \tag{3}$$

$$\alpha = (\gamma_s - 1)/2\gamma_s$$

r is the radius (or half-thickness), v - specific volume, t - time, g - heat released in the kernel per unit mass, p - pressure, u - particle velocity, a - sound speed, γ - specific heat ratio, subscript k denotes the properties of the kernel and s those of the surroundings, while superscript 0 refers to initial conditions.

As a consequence of assumptions (2) and (3) one has

$$r_k = r_i, \quad u_{is} = u_{ik} = dr_i/dt, \quad p_{is} = p_{ik} \tag{4}$$

where subscript i denotes conditions at the interface.

In terms of the nondimensional parameters

$$P \equiv p_k/p_k^0; \quad v \equiv v_k/v_k^0; \quad Q \equiv g/p_k^0 v_k^0; \quad \tau \equiv t/\delta_\omega$$

where δ_ω is the rise time of the exothermic power pulse, that is the time interval between the start of the pulse and the moment it attains its peak level, Eq. (2) becomes

$$\gamma_k P\, dv/d\tau + v\, dP/d\tau = (\gamma_k - 1)\, dQ/d\tau \tag{5}$$

while Eqs. (1), (3) and (4) yield

$$dv/d\tau = [2/\lambda(\gamma_s - 1)](P_k^\alpha - 1) \tag{6}$$

where

$$\lambda \equiv r_k^0/a_s^0\delta_\omega \tag{7}$$

appears as the appropriate Damköhler number for our problem.

Substituting Eq. (6) and its integral

$$v = 1 + \frac{}{\lambda(\gamma_s - 1)}\int_0^\tau [P^\alpha(x) - 1]\, dx \tag{8}$$

into Eq. (5), one gets the following single differentio-integral equation

$$\lambda\frac{dP}{d\tau} + \frac{2}{\gamma_s - 1}\frac{dP}{d\tau}\int_0^\tau [P^\alpha(x) - 1]\, dx + \frac{2\gamma_k}{\gamma_s - 1}P(P^\alpha - 1) = \lambda(\gamma_k - 1)\frac{dQ}{d\tau} \tag{9}$$

This can be integrated, yielding the following expression for the pressure pulse

$$P(\tau) = \frac{\lambda[(\gamma_k - 1)Q(\tau) + 1] + 2\dfrac{\gamma_k - 1}{\gamma_s - 1}\left[\displaystyle\int_0^\tau P(x)\,dx - \int_0^\tau P^{\alpha+1}(x)\,dx\right]}{\lambda + \dfrac{2}{\gamma_s - 1}\left[\displaystyle\int_0^\tau P^\alpha(x)\,dx - \tau\right]} \tag{10}$$

One obtains thus, in effect, an integral equation for $P(\tau)$ which, for the initial value problem at hand, can be numerically integrated in a straightforward manner to give the pressure pulse corresponding to a given heat release profile $Q(\tau)$. The equation can be considered, therefore, as a transfer function for the exo-exothermic reaction center where $Q(\tau)$ is the forcing term while $P(\tau)$ is the response. As it is evident from the form of Eq. (10, the primary parameter governing the dynamic behavior of the system is the Damköhler number, λ.

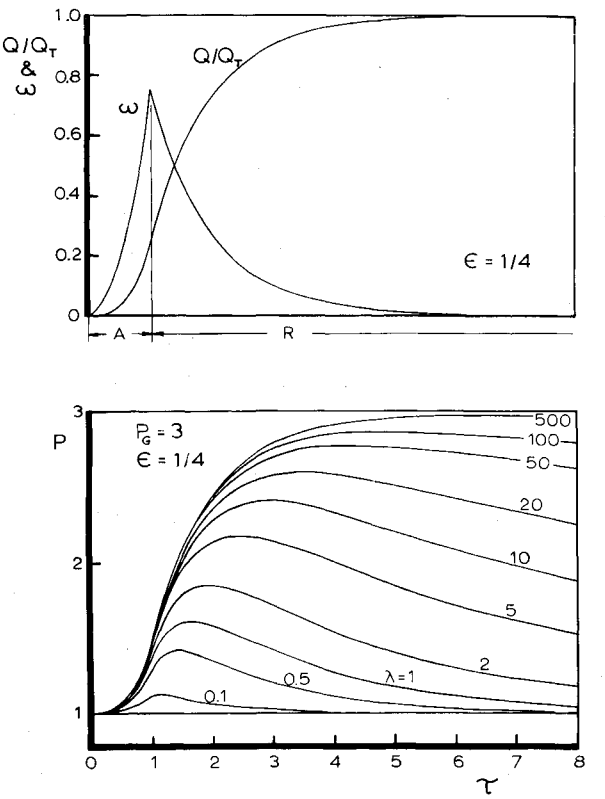

Fig. 1 Time profiles of the exothermic heat release and the corresponding power pulse specifying the input to the plane-symmetrical reaction center and the resulting pressure profiles representing the output. The excitation and relaxation zones of the power pulse are labeled *A* and *R*, respectively. Pressure profiles correspond to the case of $P_G = 3$ and $\varepsilon = \frac{1}{4}$ with $\gamma_s = \gamma_k = 1.3$, each curve being specified in terms of the Damköhler number $\lambda \equiv \gamma_k^\circ/(\alpha_s^\circ\delta_\omega)$.

Forcing Function

For the purpose of numerical analysis, the exothermic driving function, i.e. the time profile of the heat released per unit mass, is expressed in terms of the following typical form:

$$\frac{Q}{Q_T} = \begin{cases} \varepsilon\tau^2\exp(\tau - 1) & \text{for } 0 \leqq \tau \leqq 1 \\ 1 - (1 - \varepsilon)\exp[\{3\varepsilon/(1 - \varepsilon)\}(1 - \tau)] & \text{for } \tau \geqq 1 \end{cases} \tag{11}$$

The corresponding power pulse is then

$$\omega \equiv \frac{1}{Q_T}\frac{dQ}{d\tau} = \begin{cases} \varepsilon(2 + \tau)\tau\exp(\tau - 1) & \text{for } 0 \leqq \tau \leqq 1 \\ 3\varepsilon\exp[\{3\varepsilon/(1 - \varepsilon)\}(1 - \tau)] & \text{for } \tau \geqq 1 \end{cases} \tag{12}$$

In the above Q_T represents the heat of reaction per unit mass and ε is the fraction of the heat released during the rise time of the power pulse—a parameter controlling the shape of its profile. The rise time of the power pulse can be referred to as the excitation period, while its decay time corresponds to the relaxation period. Thus ε expresses the fraction of the heat of reaction released during the excitation period. The width of the pulse at one half its maximum power, $\delta_{\omega t2}$, is a measure of its sharpness and is related to both the rise time δ_ω and ε. For the class of profiles represented by Eq. (12) one has, for instance,

$$\delta_{\omega t2} \approx [0.3 + 0.7(1 - \varepsilon)/3\varepsilon]\,\delta_\omega \tag{13}$$

The functions specified by Eqs. (11) and (12) are shown in the upper part of Fig. 1 corresponding to a specific case of $\varepsilon = \frac{1}{4}$. In the analysis Q_T is treated as a parameter. Its value is expressed conveniently in terms of P_G, the pressure ratio attained by combustion at constant volume in a perfect gas system of reactants for which Q_T is the Hugoniot constant. It follows then from the Hugoniot equation[7] that

$$Q_T = (P_G - 1)/(\gamma_k - 1) \tag{14}$$

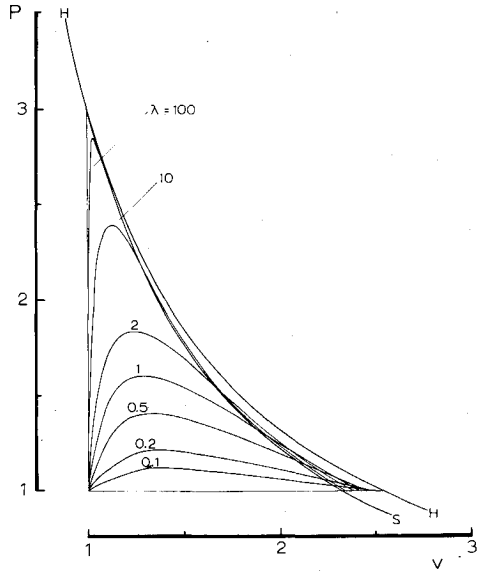

Fig. 2 Thermodynamic process paths on the plane of pressure and specific volume corresponding to the solutions given in Fig. 1. The Hugoniot curve for $P_G = 3$ ($\gamma_k = 1.3$) and the isentrope passing through the point $P = 3$, $v = 1$ are denoted as H and S, respectively.

Response

The lower part of Fig. 1 represents a set of pressure profiles obtained from Eq. (10) by numerical computations corresponding to $Q(\tau)$ given by Eq. (11) for $P_G = 3$ and $\varepsilon = \frac{1}{4}$ with $\gamma_k = \gamma_s = 1.3$. Each curve of the set is specified in terms of the value of the Damköhler number, λ.

The complementary information on the thermodynamic process paths on the plane of pressure vs specific volume is given by Fig. 2. The diagram demonstrates clearly the effect of the Damkohler number on the behavior of the system. At one extreme $\lambda = 0$ and the process occurs at constant pressure, at the other $\lambda = \infty$ and it is one of heat addition at constant volume followed by an isentropic expansion down to the initial pressure.

Peaks of pressure profiles expressed in a normalized, nondimensional form are shown plotted vs the Damköhler number of Fig. 3, with P_G appearing as a parameter, while Fig. 4 describes the corresponding variation of the rise time ratio, δ_p/δ_ω, representing the relative position in time of the pressure peak with respect to that of the power pulse of energy release. Figure 5 completes the description of the response of the reaction center for $\varepsilon = \frac{1}{4}$. It describes the dependence of the duration of the pressure pulse, expressed in terms of its time width at one half maximum, on the Damköhler number. In all cases the pressure

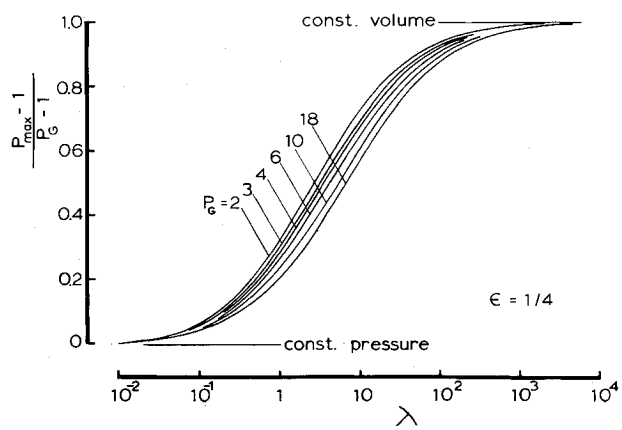

Fig. 3 Peak overpressure of the reaction center as a function of the Damköhler number with the heat of reaction per unit mass, expressed in terms of P_G, as a parameter.

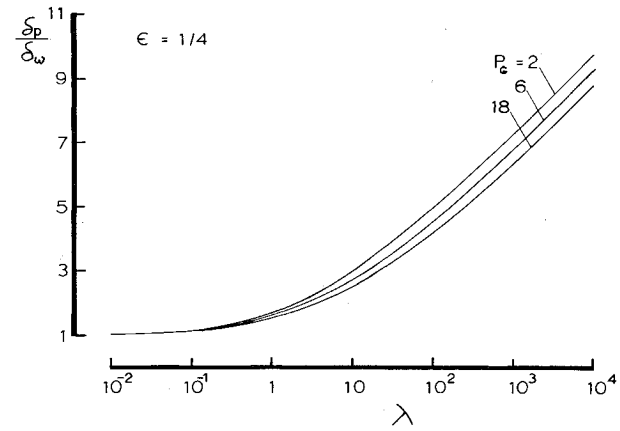

Fig. 4 Ratio of rise times of the pressure and power pulses as a function of the Damköhler number with the heat of reaction per unit mass, expressed in terms of P_G, as a parameter.

rise lags behind the power pulse, its magnitude being primarily dependent on the Damköhler number, while remaining relatively insensitive to the heat of reaction per unit mass represented by P_G.

Finally, Fig. 6 demonstrates the variation of the peak pressure and of the ratio of the rise time of the pressure and power pulse as a function of the shape parameter of the heat release profile, ε, for a given value of λ and P_G. The graph brings forth the importance of the relative duration of the excitation period—the time during which the rate of energy release increases—on the dynamic response of the exothermic reaction center.

Physical Example

To illustrate practical significance of the concepts introduced in this paper, let us consider the onset of detonation in an explosive gas mixture.

It has been demonstrated some time ago[8] that the transition to detonation is triggered in this case by a blast wave, the so-called "explosion," whose sudden appearance cannot be explained on the basis of just the gasdynamic compression ahead of the accelerating flame.[9] Its information must be therefore due to some processes occurring at the flame front which at that time is highly turbulent.

Cinematographic laser-schlieren records of the events leading to the onset of such a blast wave in a stoichiometric hydrogen-oxygen mixture which was initially maintained at a pressure of 0.915 atm and a temperature of 298° in a $1 \times 1\frac{1}{2}$ in. cross section tube are shown in Fig. 7.

Thick black traces on the schlieren records mark the position of the bulk of the turbulent flame front that, over an appreciable

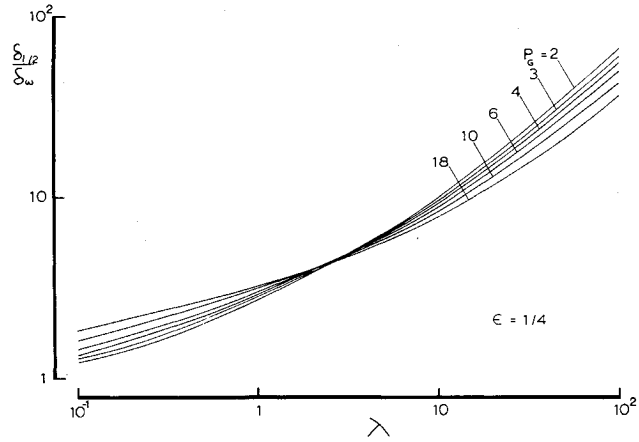

Fig. 5 Pressure pulse half-width (time width at $\frac{1}{2}$ maximum overpressure) as a function of the Damköhler number with the heat of reaction per unit mass, expressed in terms of P_G, as a parameter.

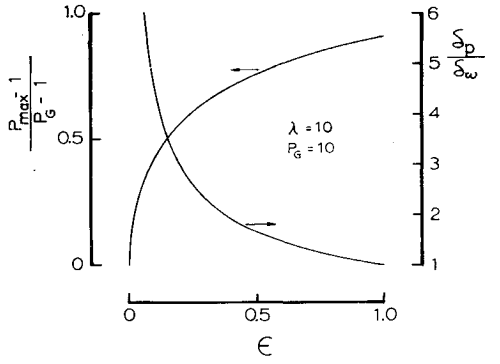

Fig. 6 Peak overpressure of the reaction center and the corresponding rise time ratio as a function of the power pulse shape parameter, ε, for the case of $\lambda = 10$ and $P_G = 10$.

extent in optical depth, is parallel to the optical axis. As it appears there, the onset of the blast wave is evidently associated with the rapid convergence between such two opposite flame fronts. One may just speculate that the blast wave was formed as a consequence of a collision between two flame fronts. On the basis of our theory one can estimate the strength of such a blast wave expressed in terms of the overpressure it can generate.

For this purpose one has to assume that the flame fronts are locally plane and parallel to each other—a plausible idealization in view of the records of Fig. 7. Under such circumstances the total time for energy release in a kernel of initial radius r_k^0 that is consumed by colliding flames whose burning speed is on the average S_f is r_k^0/S_f. The corresponding excitation time is then

$$\delta_\omega = \varepsilon r_k^0/S_f \tag{15}$$

and, hence, according to Eq. (7), the Damköhler number is

$$\lambda = S_f/\varepsilon a_s^0 \tag{16}$$

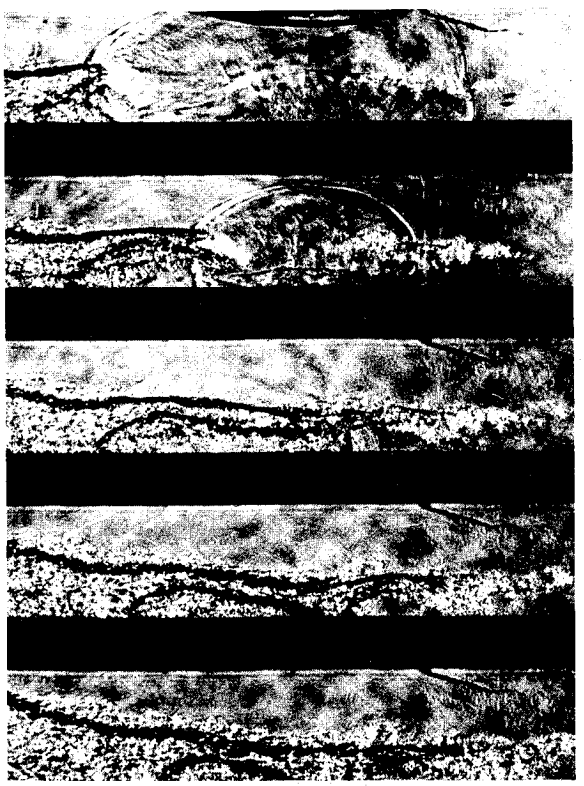

Fig. 7 Cinematographic laser-schlieren record of the transition to detonation in a spark-ignited $2H_2 + O_2$ mixture contained in a duct of $1 \times 1\frac{1}{2}$ in. cross section, initially at a pressure of 0.915 atm and a temperature of 298°K. Framing interval: 5 μsec; width of test section: 1 in.

From the analysis of the nonsteady gasdynamic processes that occurred in the unreacted gas mixture prior to the onset of the blast wave,[9] it was deduced that the flame propagates then into a medium at a pressure of 14.3 atm and a temperature of 640°K, where the local velocity of sound is 780 m/sec, while the particle velocity is 1450 m/sec. The equilibrium Hugoniot conditions with respect to such an initial state of the hydrogen-oxygen mixture correspond to $P_G = 4.94$, while the velocity of sound in the burned gas is $a_f = 1500$ m/sec. The gasdynamic analysis of the schlieren records yielded also for the relative flame speed a constant value of $S_r = 200$ m/sec.

Since the pressure wave generated by an exothermic reaction center created by flame collision propagates into the burned gas we have $a_s^0 = a_f$. At the same time, neglecting the accelerating effect on the flame speed of the overlap between the preparation zones that must have taken place just prior to the collision, we can identify S_f with S_r. From the analysis of the dynamics of the plane exothermic reaction center governed by the reaction kinetics of the hydrogen-oxygen system we have obtained for ε the value of 0.25.[1] Thus from Eq. (16) one gets for the Damköhler number $\lambda = 0.533$.

Figure 3 yields then

$$(P_{max} - 1)/(P_G - 1) = 0.19$$

whence

$$P_{max} = 1.75$$

Considering the fact that this represents the effect of a single reaction center, while the reacting medium is affected by a certain number density of coherent centers,[2] the above result should be certainly of sufficient significance in providing a rationale for the onset of the blast wave observed on Fig. 7.

Conclusions

The analysis presented here of the dynamic behavior of an idealized, plane-symmetrical exothermic reaction center leads to the following conclusions:

1) The theory brings forth the significance of considering the exothermic process as one consisting of a so-called here excitation period followed by a relaxation period; the first is defined as the process associated with an increasing rate of heat release (or exothermic power pulse) while during the second this rate decreases. The above distinction has, to our knowledge, never been made in the analysis of exothermic processes, while, as it appears from our studies, it ought to be of definite advantage in assessing the dynamic properties of the reacting system.

2) The macroscopic parameters of the excitation process are: a) the Damköhler number $\lambda \equiv r_k^0/a_s^0\delta_\omega$ where r_k^0 is the initial radius of the kernel of the reaction center, a_s^0 the initial sound speed in the surroundings, and δ_ω the excitation period. b) the fraction of heat released during the excitation period $\varepsilon \equiv g_E/g_T$ where g_T is the heat of reaction per unit mass.

3) The Damköhler number of zero corresponds to a constant pressure process, while infinity yields a process of heat addition at constant volume followed by an isentropic expansion back to the original pressure. The plot of maximum overpressure generated by the reaction center, nondimensionalized with respect to P_G - the equilibrium pressure attained by heat addition at constant volume, i.e. $(P_{max} - 1)/(P_G - 1)$ is a sensitive function of the Damköhler number λ in the range of from $\lambda = 10^{-2}$ to $\lambda = 10^4$ (for $\varepsilon = \frac{1}{4}$) while being relatively insensitive to P_G expressing, in effect, the heat of reaction.

4) The ratio of the rise time of the pressure pulse to the excitation period, are similarly dependent much more on the Damköhler number than on the heat of reaction.

5) The effect of the fraction of the heat of reaction released during the excitation period, ε, is most significant to both the maximum over-pressure and to the ratio of the rise time of the pressure pulse to that of the power pulse; the increase of ε from 0 to 1 for a constant value of the Damköhler number and the heat of reaction being associated with a similar change in

$(P_{max} - 1)/(P_G - 1)$ from 0 to 1 while δ_p/δ_ω decrease from ∞ to 1, the high overpressure corresponding to a sharp pressure pulse.

6) Application of the theory presented here to the problem of the transition to detonation provides a reasonable explanation for the generation of the blast waves that triggers this process.

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End Effects in Faraday Type MHD Generators with Nonequilibrium Plasmas

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The influence of the end losses on a linear nonequilibrium Faraday type MHD generator with finite segmented electrodes is studied theoretically. Numerical two-dimensional calculations are performed for constant, parabolically increasing and parabolically decreasing magnetic induction near the generator exit. The plasma considered consists of argon at 5 atm and 2000°K and seeded with 0.2% cesium. The influence of the described magnetic induction configurations on the current distribution in the MHD-generator exit is studied. The effect of several configurations on the existence of a region near the generator exit with higher electrical conductivity than in the generator itself is discussed. It is preferable to extend the magnetic induction over a distance equal to the generator height, keeping the value of the magnetic induction constant; then the electrical conductivity is not increased near the generator exit and the homogeneous current pattern is hardly disturbed. From the given analysis information is obtained for the design of an appropriate magnetic induction configuration.

Introduction

At the generator ends, because the magnetic induction varies as a function of the distance along the MHD generator, additional forces work on the plasma streaming through the generator whereas such forces are not present in the central region. These forces disturb the homogeneous current distribution in the end regions. In the generator the electrical conductivity of the plasma is reduced roughly by the factor $1/(1 + \beta^2)$ relative to that in the end regions. Owing to the eddy currents, whose existence will be deduced, the electrical conductivity at the generator exit may increase further. This conductivity variation has been experimentally verified by Zauderer.[1] In nonequilibrium MHD generators, the measured open-circuit voltages show significantly lower values than the theoretical values.[2,3]

Near the generator ends these voltages are still further reduced. Decher[2] explains the open-circuit voltages by means of a model in which the end regions are electrically coupled to the coreflow by electrode wall layers. Further, experiments have shown that axial end currents produce reversal of the axial electric field.

To reduce end effects, Sutton and Sherman[4] install isolating vanes in the flow. Another suggestion to diminish the electrical conductivity of the plasma is to increase its recombination rate near the generator exit by the injection of water into the plasma.[5] End losses have been calculated by Dzung[6] and Sutton.[7] In their one-dimensional analysis for an infinitely segmented Faraday generator, the electrical conductivity σ and the Hall parameter β are supposed to be constant.

This paper reports a two-dimensional analysis of the end effects for a finitely segmented Faraday generator as a function of the magnetic induction configuration. In the mathematical model allowance is made for nonequilibrium ionization and relaxation effects in electron temperature and electron density.

The numerical calculations are performed for constant current per electrode, and this because the maximum generator output and the maximum efficiency can be obtained in a constant current density generator.[8] No periodic field distributions have been assumed. Three configurations of the magnetic induction (one constant, one parabolically increasing and one parabolically decreasing) are used in the calculations. The method of solution is similar to Blom's.[9] All calculations have been carried out for an argon plasma seeded with 0.2% cesium, at 5 atm, a heavy particle temperature of 2000°K, and Mach number 1.38. These conditions were chosen because they exist in the Eindhoven shocktube experiment, the MHD generator of which has an outlet cross section of 0.1×0.09 m^2.

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Index category: Plasma Dynamics and MHD.

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