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A FUNDAMENTAL MATHEMATICAL THEORY FOR THERMAL EXPLOSIONS IN RIG--ETC(U)

JUL 82 J BEBERNES, D R KASSOY

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A study has been made of the evolution of spatially variable thermal explosion
processes occurring in finite systems. Mathematical models have been developed
for both rigid systems and for combustible, compressible gas mixtures. Purely
thermal processes occur in the former. Gasdynamic events play a significant role
in the latter. Solution development is carried out in terms of asymptotic
expansions valid for the limit of high activation energy. Physical processes
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occurring on disparate time and length scales are systematically delineated. Spontaneously ignited reaction processes are shown to evolve into highly localized hot spots in both rigid and gaseous systems. In the latter case gas expansion processes cause a mechanical response in the system that leads to acoustic fields and significant gasdynamical processes. Detailed solutions are presented for symmetrical systems (slabs, cylinders and spheres). More general systems are studied by using qualitative analytical tools to ascertain fundamental solution properties like bounds on trajectories, estimates of escape time (thermal runaway) and effects of compressibility on thermal runaway.

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A FUNDAMENTAL MATHEMATICAL THEORY FOR
THERMAL EXPLOSIONS IN RIGID SOLIDS AND IN GASES

Final Report

CUCOMB 1

J. Bebernes and D.R. Kassoy

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I. Introduction

A study has been made of the evolution of spatially variable thermal explosion processes occurring in finite systems. A one-step global reaction involving the oxidation of a fuel to products is assumed to occur in a rigid or in a gaseous combustible mixture. The exothermic reaction is described by an Arrhenius rate law in which the activation energy is large relative to the internal energy of the system. In general the combustible is assumed to be in an equilibrium state initially. The thermal reaction is autoignited at the relatively low initial temperature in all cases studied. In addition a wall temperature disturbance, prescribed in some circumstances, is used to alter the explosion development.

Much of the mathematical modelling has been developed for processes in a rigid explosive material. In this case the time-history of the thermal reaction is a result of pointwise competition between chemically generated heat addition and heat transfer due to conduction. The distribution of reactant is determined solely by chemical conversion processes. Species diffusion is neglected. The explosive material does not respond in any manner to the sometimes dramatic thermal and chemical events occurring within. For example, thermal expansion of the explosive, the concomitant mechanical response, possible material failure (fracturing) and gaseous product generation are ignored. As a result the theory for rigid materials has a limited application.

The earliest significant development in a rigid explosive occurs on the conduction timescale of the container. During most of the induction period the temperature deviation from the initial state is small. However, when a critical explosion time is approached a thermal runaway occurs as the deviation becomes unbounded. This singularity occurs in a highly localized spatial region. In a subsequent time period of extremely brief duration the temperature (concentration)

increases (decreases) significantly in the delimited hot spot zone where fast reaction processes dominate the physics. Elsewhere, the system is nearly invariant because physical processes are controlled basically by relatively slow conduction phenomena. In principle the hot spot can act as an ignition source for a classical low speed flame (A.K. Kapila, SIAM Applied Math. 39, pp. 21-36, 1980). However, it appears likely in practice that the thermo-mechanical events described earlier would have a significant influence on further evolution of the combustion event.

Theoretical consideration of an explosive gas mixture must include thermo-mechanical processes associated with gas compressibility, acoustic propagation and stronger gasdynamical phenomena (e.g. shocks) that are characterized by the short acoustic time scale of the vessel. In contrast reaction may occur on the long conduction time scale when the temperature is near the initial value but also on a much shorter time scale as the temperature rises. Of course conductive heat transfer is always limited to the conduction time. The initial induction period is much like that for the rigid explosive. However, gas compression due to confined heating causes additional (compression) heating and a shortening of the explosion time. In addition spatially differential expansion rates are responsible for acoustic wave generation. The subsequent hot spot formation in a compressible gas involves gasdynamical processes as well as purely thermal events. In particular the appearance of nonlinear acoustic waves, weak shocks and their possible evolution into detonation waves must be considered. It is apparent that there will be a far richer panoply of physical processes in the gaseous explosion than those occurring in a rigid material.

Solution development has been based on a variety of perturbation methods employing the limit of high activation energy. These techniques permit one to delineate processes occurring on rather disparate length and time scales. While

a single parameter suffices to define the rigid explosive problem, the gaseous case requires a two parameter expansion. The second parameter is basically the ratio of the acoustic to conduction times in the vessel.

While one may pose initial-boundary value problems which are amenable to explicit solution development there are others where only a qualitative treatment is feasible (short of large scale numerical computation). Problems involving generalized initial and boundary conditions and non-symmetric geometries are in the latter category. Thus a major emphasis has been made to develop quantitative analytical tools for assessing the general solution properties of the describing equations.

For example, the induction model for a reactive gas in a bounded container required especially sharp analytical tools to qualitatively determine precisely the solution behavior. This particular problem was resolved using a sophisticated semigroup analysis and comparison techniques.

The mathematical tools which were developed to deal with time-varying thermal explosions have a much wider applicability. For example the evolution of a spatially homogeneous model of coal particle combustion can be described in terms of high activation energy asymptotic solutions. The periods of inert heatup, ignition, significant burning and extinction can be delineated in a systematic, lucid manner.

II. Description of Significant Results

Each of the manuscripts developed by the research personnel is described in some detail below.

1. D.R. Kassoy and J. Poland, "The Thermal Explosion Confined by a Constant Temperature Boundary: II - The Extremely Rapid Transient", SIAM J. Appl. Math., 41, pp. 231-246 (1981).

The extremely rapid transient phase of a thermal explosion in a vessel with constant wall temperature is described. The solution is developed in

terms of a nearly-inert, cool, reactant-rich conduction-controlled zone and a highly localized veritable fireball controlled by chemical reaction rates, which are separated by a distinct zone of time-invariant spatial structure. In the first region the temperature is only slightly different from the initial value. In contrast, the variation in temperature in the fireball is large, ranging almost to the adiabatic explosion value. The rise in temperature is accompanied by significant fuel consumption. When the fireball formation is nearly complete, the fuel available therein is vanishingly small. The theoretical formulation provides a specific time scale for the rapid transient process, which is extremely short in comparison to that in the induction period which precedes the explosive event. The maximum temperature T_M in the fireball is 0(1) less than adiabatic explosion value and is dependent on the geometry of the system. For a given set of physicochemical parameters the value of T_M is largest in a slab and smallest in a sphere relative to the intermediate value in a cylinder. This result can be explained in physical terms by recognizing that for a given volume of combustible the surface area for heat loss is minimized in a slab and maximized in a sphere. It is concluded that for equal heat release rates per unit volume the explosive event is most dramatic in a slab and least dramatic in a sphere.

The fireball size, found to depend on the material properties of the combustible, is extremely sensitive to the activation energy of the one-step reaction mode. As shown in Table 1 a decrease in the activation energy parameter ϵ from 5×10^{-2} to 2×10^{-2} causes a reduction of 10^{-5} in the magnitude of the fireball size. Fireballs are larger in spherical geometry than in a slab configuration.

TABLE 1
Fireball Size

Frank-Kamenetskii parameter value	$\delta = 2.5$	$\delta = 20$
ϵ	$r'_{\infty}(\text{cm.})$	$r'_{\infty}(\text{cm.})$
.05	7.26×10^{-1}	6.19×10^{-1}
.04	1.00×10^{-1}	8.20×10^{-2}
.03	3.70×10^{-3}	2.83×10^{-3}
.02	5.04×10^{-6}	3.38×10^{-6}

2. J. Bebernes and D. Kassoy, "A Mathematical analysis of blow up for thermal reactions -- the spatially nonhomogeneous case", SIAM J. Appl. Math., 40, 576-584 (1981).

The equations describing the induction period process for a supercritical, high-activation energy thermal explosion in a bounded domain are studied. A formal proof, based on comparison techniques, is used to show that the temperature perturbation becomes unbounded as $t \rightarrow t_B \leq \infty$ for values of the Frank-Kamenetski parameter, δ , greater than the critical value. Upper and lower bound estimates for t_B are found by using specified comparison equations. A comparison of these bounds with values of t_B obtained from numerical solutions of the basic equations shows that the estimates provide an excellent prediction of the escape time when δ is greater than 2-3 times the critical value. For smaller values of δ , where heat loss is more significant, a better comparison equation is required.

3. J. Bebernes, "A Mathematical Analysis of Some Problems from Combustion Theory", Quaderni dei Gruppi di Ricerca Matematica del CNR, 1981, 63 pp.

This monograph presents a detailed mathematical analysis of several specific models which occur in combustion theory.

4. J. Poland, I.O. Hindash and D.R. Kassoy, "Ignition Processes in Confined Thermal Explosions", *Combustion Science and Technology*, 27, pp. 215-227 (1982).

The induction period of a high activation energy thermal explosion in a confined, rigid, nondiffusing, combustible material is considered. A method-of-lines approach is used to develop numerical solutions for the infinite slab, infinite cylinder and spherical geometries. When the dimensional initial and boundary temperatures are equal, results are obtained for a wide range of supercritical values of the Frank-Kamenetskii parameter δ , and for arbitrary values of the ratio of the chemical heat release and the initial internal energy. The time-history of the spatially variable temperature distribution is described. At a common value of δ the thermal runaway time is largest for a sphere and smallest for a slab, with the cylindrical system between as shown in Fig. 1. The explosion location r_e is found to move away from the symmetry point when the wall temperature disturbance is made sufficiently large compared to the initial temperature. A further increase in the former moves the value of r_e toward the hot boundary, as shown in Fig. 2.

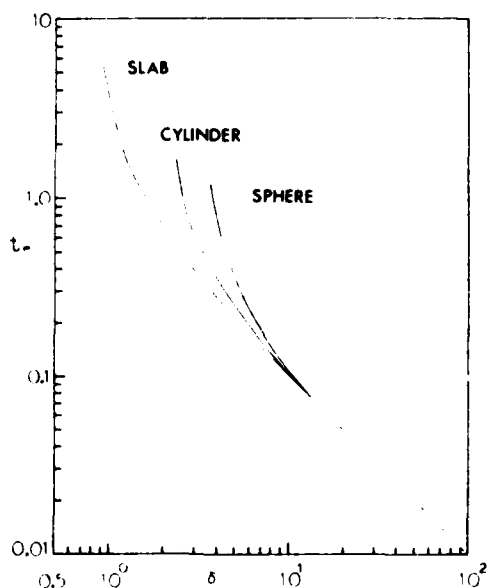


Figure 1 The explosion time t_e versus δ for the slab, cylinder and sphere.

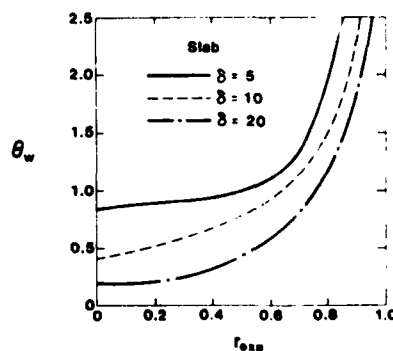


Figure 2 The wall temperature disturbance θ_w as a function of the explosion location for a slab at $\delta = 5, 10, 20$.

The dependence of the explosion time on the incremental difference between the boundary and initial temperatures and on a modified Frank-Kamenetskii parameter is described in detail. The time-history of an off-center explosion is shown to produce precisely the same kind of highly localized hot spot as that found when the temperature difference is zero.

5. J. Bebernes and A. Bressan, "Thermal behavior for a confined reactive gas", J. Differential Equations, 44, 118-133 (1982).

Using a high activation energy asymptotic analysis on the complete describing equations for a compressible reactive gas, Kassoy and Poland derived the following induction model:

$$(G) \quad \left\{ \begin{array}{l} \theta_t - \Delta \theta = \delta e^{\theta} + \frac{\gamma-1}{\gamma} \cdot \frac{1}{\text{vol} \Omega} \int_{\Omega} \theta_t dy \\ \theta(x,0) = \theta_0(x), \quad x \in \Omega \\ \theta(x,t) = 0, \quad x \in \partial \Omega, t > 0 \end{array} \right.$$

Using semigroup techniques, we are able to give a precise time-history description for the temperature perturbation $\theta(x,t)$. Existence, uniqueness, and continuous dependence of solutions are proven for general n -dimensional domains Ω . For spherical domains, further qualitative results are provided, concerning the global behavior of solutions and the existence of a finite blow-up time for $\delta > \delta^*$. In particular, we show that the temperature perturbation for an ideal gas is always greater than that for a solid fuel in identical bounded containers and hence a gas explodes sooner than a solid fuel. Physically, this can be explained by the additional generation of heat due to the compression of the gas.

6. J. Poland and D.R. Kassoy, "The Thermal Explosion in a Confined Reactive Gas. I - The Induction Period Solution", to appear Combustion and Flame (1982).

The initiation of gasdynamical processes during the induction period of a high activation energy supercritical thermal explosion in a confined reactive gas is modelled mathematically. Basically we find that the induction period is shortened relative to an analogous process in a rigid explosive because compression heating of the constant volume system accelerates the reaction process. The detailed development of the density, velocity, temperature, and fuel mass fraction fields during the induction period is separated into three phases. During the first phase, occurring on the acoustic time scale of the vessel, conduction dominated boundary layers adjacent to the cold wall generate an acoustic field in a non-dissipative interior core region. The second phase, occurring on the conduction time scale of the vessel, is characterized by a pointwise competition between reaction generated heat release, conduction, and compression. The Frank-Kamenetskii criteria dividing super- and subcritical systems is found to be the same as that for rigid explosive materials. In a supercritical system, $\delta > \delta_{crit}$, the third phase, of extremely limited duration, is dominated by the development of a tiny self-focusing hot spot embedded within a nearly invariant conduction-dominated field filling most of the vessel. The rapid gas expansion in the hot spot is the source of further, more dramatic gasdynamical processes.

7. D.R. Kassoy and Paul A. Libby, "Activation Energy Asymptotics Applied to Burning Carbon Particles", in press, Combustion and Flame (1982), also MRC Technical Summary Report #2322, January 1982, Mathematics Research Center, University of Wisconsin-Madison, 610 Walnut Street, Madison, Wisconsin 53706.

The method of activation energy asymptotics is used to describe the entire history of a cold carbon particle immersed suddenly in a hot oxidizing ambient which may involve oxygen but perhaps other active species such as carbon dioxide

and water as well. The history of such a particle involves an initial, pre-ignition period during which the particle is heated but undergoes no significant chemical reaction. When the ambient temperature is sufficiently high, the particle reaches a critical, ignition temperature and heterogeneous reactions involving the attack of carbon becomes effective. The ignition period is followed by post-ignition behavior involving rapid increases in particle temperature which, as a consequence of chemical reaction, generally exceeds that in the ambient. Under the circumstances of practical interest and assumed to prevail in this study both the ignition and post-ignition periods are brief but result in the rate of mass loss by the particle becoming diffusion-limited. There ensues an extended period involving a constant rate of mass loss and terminating in a complex, although brief extinction period during which consumption is complete. The brevity of the transition periods associated with ignition and extinction implies that the principal features of particle behavior we consider are given by a simplified analysis based on inert particle heat-up and diffusion-limited combustion.

The asymptotic analysis provides a description of the particle time history which is quite like that found by using an approximate theory based on ad hoc assumptions. However, the present analysis provides clear indications of the conditions under which the analysis applies. In fact a significant portion of our discussion relates to the applicability of a specific numerical calculation involving ϵ small compared to unity but nonzero to the limiting behavior arising when the small parameter $\epsilon \rightarrow 0$. For example, during a brief extinction period all of the quantities describing particle behavior undergo significant variations. As a consequence the asymptotic solutions for this period are complex and impose significant limitations on the acceptable size of the expansion parameter. In fact only two of the four zones found to be

required are compared successfully to an exact numerical solution.

The asymptotic analysis provides estimates for quantities of applied interest: conditions under which ignition occurs, conditions for diffusion-limited mass loss and the times for ignition and complete consumption.

8. J. Bebernes and R. Ely, "Comparison techniques and the method of lines for a parabolic functional equation", Rocky Mountain J. Math., 12, 723-733 (1982).

The temperature perturbation solution $\theta(x,t)$ of the induction period model for a reactive gas in a bounded container Ω given by

$$(G) \quad \left\{ \begin{array}{l} \theta_t - \Delta \theta = \delta e^{\theta} + \frac{\gamma-1}{\gamma} \frac{1}{\text{vol} \Omega} \int_{\Omega} \theta_t \, dy \\ \theta(x,0) = \theta_0(x) \, , \quad x \in \Omega \\ \theta(x,t) = 0 \, , \quad (x,t) \in \partial \Omega \times (0,\infty) \end{array} \right.$$

is dominated by the solution $u(x,t)$ of

$$(E) \quad u_t - \Delta u = \delta e^u + \frac{\gamma-1}{\text{vol} \Omega} \cdot \delta \cdot \int_{\Omega} e^u \, dy$$

on their common interval of existence. In this paper an extensive analysis is given for the class of explicit integro-partial differential equations of the form

$$(D) \quad u_t - \Delta u = f(t,u) + \int_{\Omega} g(t,u) \, dx$$

which include (E) as a special case. In particular, we show that the method of lines can be used to construct solutions to approximating systems of ordinary differential equations which converge to the solution $u(x,t)$ of (D).

9. D.R. Kassoy, "A Note on Asymptotic Methods for Jump Phenomena", to appear SIAM J. Applied Math. (1982).

Traditional singular perturbation methods are employed to develop a solution to the differential equation $y' = y^2(1-y)$, $y(0) = \epsilon \ll 1$ considered by Reiss (SIAM J. Applied Math. 39, pp. 440 - 455, 1980), which models an elementary chemical process. The results are compared with those found by Reiss who used a novel asymptotic method to construct solutions which exhibit rapid transient behavior. It is shown that Reiss' jump solution corresponds to the asymptotic (large time) representation of the more complete solution found from a formal matched asymptotic expansion procedure. A comparison of results in the rapid transition region obtained from the exact solution, from those found by Reiss and from the matched asymptotic expansion solution developed here, show that the last is far more accurate than the second. Since the systematically developed asymptotic expansions are far more complicated than those obtained by using Reiss' technique, one must balance the need for accuracy against the level of effort required to obtain precise results.

10. J. Bebernes and D.R. Kassoy, "Gasdynamic Aspects of Thermal Explosions", Transactions of the Twenty-Seventh Conference of Army Mathematicians, ARO Report 82-1, Army Research Office, Box 12211, Research Triangle Park, NC 27709.

In an attempt to overcome the limitations of the rigid explosive model we describe a model for the thermal explosion occurring in a confined compressible, perfect, reactive gas mixture. The problem is described by the complete equation of motion for a compressible, ideal, reactive gas mixture. All transport properties are included. Simplifications are confined to the details of the chemical reaction and material properties.

The inclusion of material compressibility in the model means that one must consider physical processes on the acoustic time scale as well as those

occurring on the conduction and reaction time scales. In particular, material expansion associated with localized heating causes mechanical disturbances in the gas. These are propagated as acoustic waves at the local speed of sound. Under appropriate circumstances the initially linear acoustic processes can become nonlinear leading to the formation of shock waves. The interaction of the shock with reactive gas introduces an entirely new set of combustion processes into the confined thermal reaction system.

A high activation energy asymptotic analysis is used as the method of solution development. Very early in the process, on the acoustic time scale of the vessel, an acoustic field is generated by spatially variable thermal expansion in the gas. The familiar induction period process is shown to develop on the conduction time scale of the vessel. Unlike previous theories for rigid materials the describing equations include effects of compressibility and deformation. The former effect is shown to cause a gaseous system to have a thermal runaway sooner than an equivalent rigid system. The effect of deformation is observed in the form of a rapidly expanding hot spot. Typical numerical solutions are presented along with theoretical considerations of the integrodifferential equation which describes the energy balance. It is shown that the basic initial-boundary value problem associated with the integrodifferential equation has a unique solution for general geometries. When the container is symmetric in n -dimensions and the initial temperature is zero, then the solution is always symmetric. Finally, it is known that the temperature for an ideal gas is always greater than that for a rigid fuel in identical bounded containers and hence a gas explodes sooner than a solid fuel. Physically, this can be explained by the additional generation of heat due to the compression of the gas.

A final-value analysis describes the thermal runaway singularity. The nonuniformity in the asymptotic expansions is shown to imply that a sequence of dramatic gas dynamic events will follow the induction period process. For example, it can be shown that the vigorous expansion rate of the hot spot edge can lead to the generation of a nonlinear acoustic signal which "breaks", thus forming a shock at a distance from the center of the hot spot which is small relative to the container size. The analysis of this process is similar to Cole's study (see Perturbation Methods in Applied Mathematics, Blaisdell (1968)) of the shock wave generated by an accelerating piston. Of fundamental interest is the shock wave strength as a function of the physico-chemical parameters of the reacting system. The details of this dependence remain to be carried out.

It is useful to speculate on the possibility that shock generation by the developing hot spot can lead to direct initiation of a detonation. If the shock is sufficiently strong the induced temperature rise will initiate a rigorous chemical reaction just behind the wave. Should the local reaction time be commensurate with the local transit time of the shock, then the reaction zone will move with the shock. And so a detonation is initiated!

11. J. Bebernes, "Ignition for a gaseous thermal reaction:", Proc. EQUADIFF V, to appear.

This paper summarizes many of the known results for the induction models for rigid and gaseous fuels in a bounded container.

III. Scientific Personnel Supported

Mithat Birkan, Ph.D. in progress,

Alberto Bressan, Ph.D. expected August, 1982,

Richard Ely, Ph.D. completed May, 1982,

I. O. Hindash, M.S. completed December, 1980.