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STUDIES IN COMBUSTION AND EXPLOSION

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A. Abstract.

The premixed flame (the self-sustained wave of exothermic chemical reaction) is the most natural mode of gaseous combustion and, hence, the main object of interest in combustion theory.

The objective of the present research is to understand and quantify the basic physical mechanisms occurring in combustion systems involving premixed gas flames and relate the acquired information to practical situations (e.g. spark-ignition engines, gas-fired burners, accidental air-fuel explosions).

Specifically, our research efforts were focused on the formulation of mathematical models for the observed novel phenomena associated with (1) effects of turbulence on burner stabilized flames, (2) transition to detonation in burner stabilized flames, (3) stability of counterflow stabilized flames, (4) self-fragmentation in near-limit flames, (5) chaotic dynamics in premixed flames, and (6) the effect of stirring on the limits of thermal explosion. The studies of combustion fundamentals of the type addressed in this project broaden the conceptual base of the pertinent engineering design fields and may be instrumental in finding new options of enhancing energy saving and production, determination of safe operating conditions and the reduction of pollution in the urban and industrial environment.

B. Table of Contents.

C. Project Output.

An extensive range of problems on flame stabilization, stability, transition of detonation and extinction/ignition limits has been studied. The details can be found in the bibliography (Sec. E). In the following we present a brief account of the results obtained.

C.1. Effects of Turbulence on Burner Stabilized Flames.

This study is motivated by new experimental studies conducted by D. Bradley (University of Leeds) which provided new evidence that for each gaseous premixture there is a certain level of turbulence at which the speed of the premixed flame reaches its maximal value. A further increase in the flow intensity leads to a drop of the flame speed, interface fragmentation, and eventual extinction of the flame.

It has been suggested that the phenomenon is likely to be controlled by the flame stretch. Such an interpretation, however, needs a qualification. The stretch indeed is known to reduce the reaction rate, provided it is positive. Otherwise, the burning will be enhanced by the stretch rather than suppressed. In turbulent flow the sign of the stretch alternates along the front. Thus it is not at all clear how the inhibiting influence of the stretch occurring in some parts of the flame will dominate the other parts of the flame and result in the overall flame extinction. Moreover, there are serious indications that, while the stretch certainly plays an important role, in order to ensure the complete extinction, some level of heat losses in indispensable. The stretch idea of flame extinction, therefore, still involves questions, the elucidation of which calls for a more fundamental approach. The principal difficulty in modeling turbulent combustion, as in many other turbulence related problems, is the wide range of spatio-temporal scales involved. It seems intuitively plausible, however, that the multiple scale nature of the flow field is not crucial for the physics of the flow induced quenching and the phenomenon may well be understood in the framework of a one-scale flame-flow interaction scheme. As has been shown in our previous (*Brailovsky & Sivashinsky, Phys. Rev. E.* 51 (1955) 1172-1183) and current studies of the problem, one may gain a good deal of relevant information even when the underlying flow is chosen as time-independent, space periodic and undirectional. In our *Phys. Rev.* (1995) paper the problem was analyzed in the framework of the so-called Slowly-Varying-Flame (SVF) formalism. The results obtained successfully reproduced the basic experimental trends, and may well-be interpreted in terms of the flow induced extinction, Figure (1). Yet some basic questions still remained unanswered. The point is that SVF formalism, by its very nature, is valid only for the flame-flow interactions where the burning velocity (flame speed relative to the underlying flow-field) is positive along the entire flame interface. The collapse of the positive velocity solutions may, however, by merely a sign of transition to non-positive velocity flames or incomplete combustion rather than local or total extinction. To gain an insight into this question the direct numerical simulation of the pertinent reaction-diffusion-advection model has been undertaken. The numerical explorations conducted for adiabatic, high Lewis number premixtures produced the following results. In the absence of heat losses there seems to be always a solution for any flow intensity (A) however high. Moreover, for the large scale flows the system becomes bistable with a hysteresic transition between stable propagation modes, Figure (2). In line with analytical predictions, extension of the analysis over non-adiabatic flames demonstrated that flame extinction by periodic shear flow is indeed feasible. It is significant that the level of heat losses required to quench the corrugated flame appears to be markedly lower than that for the planar flame. An obvious mathematical advantage of the periodic shear flow is its ability to sustain a time-independent reaction wave while bringing in the basic feature of the large-scale turbulent combustion: the flame interface extension through its corrugation. As a natural next step in imitating turbulent flows the shear flow was replaced by a more isotropic double-periodic one. In this case, apart from being distorted, the propagating flame is subject to time-periodic pulsations induced by the train of eddies crossing the flame. Even for the physically most simple Huygens model, when the flame is regarded as a geometrical interface moving at a prescribed constant burning velocity the flame dynamics becomes much more intricate than in the shear flow case. At sufficiently high stirring intensity (A), for example, the flame may eject pockets of unburned gas. The effect has also been observed experimentally by Ronney and co-workers (24^{th} *Symp. (Int.) Combust.* (1992), 543-551) in their studies of aqueous autocatalytic reaction waves crossing an array of single-scale Taylor-Couette vorticities. Note that for the double-periodic flows the SVF formalism, due to the emergence of pathological time-dependent solutions becomes largely ineffective. Thus, to describe the negative velocity flames as well as the flame extinction both Huygens and SVF formulations are inadequate and here again to gain an insight into the flame-flow interaction direct numerical simulations were involved. Figure (3) shows some results obtained for the double periodic flow in the absence of heat losses.

For the chosen range of the flow length-scales the flame speed (V) response to stirring intensity (A) appears to be somewhat different from the shear flow case. Here with the stirring strengthening the flame speed goes over a maximum and thereupon settles at some level independent of A. Such a striking outcome may be explained by the analysis similar to that developed in our recent study of the effect of stirring on the limits of thermal explosion (*Kagan, Sivashinsky et al. Combust. Theory & Modelling*, 1 (1997), 97-111). The point is that $A \gg 1$ the isotherms and isoconcentrations tend to coincide with the flow streamlines thereby strongly affecting the inner flame structure and hence its local burning velocity. The resulting dynamical picture thus will markedly differ fro the Huygens model where the flame structure is virtually unaffected by the underlying flow. Similarly to the shear flow case but in a much more pronounced manner the incorporation of mild heat losses results in the flame extinction provided the stirring intensity is high enough (Figure 4). The evaluated flame speed response to the stirring intensity is found to be in excellent qualitative correlation with recent experiments on flame extinction by turbulence. These studies are reported in the papers [1,2] (Sec. E).

C.2 Transition to Detonation in Burner Stabilized Flames.

New experimental studies with the pulsed flame detector conducted by S. Cheskis (Tel Aviv University) showed that under certain conditions the well settled pulsating combustion is accompanied by rather strong claps suggesting transition from conventional subsonic regime to detonation. The pulsed flame burner, apart from its other functions, thus unexpectedly appears to be a natural system for studying deflagration-to-detonation transition, one of the central and still poorly described issues of gaseous combustion. Normally, deflagrations are initiated by a mild energy discharge, e.g. by a spark, while detonations are provoked by shock waves via localized explosion. A shockless initiation of detonation is also feasible. It may occur in appropriately preconditioned, e.g. non-uniformly preheated gas. Thus, both deflagration and detonation appear to be stable attractors each being linked to their own base of initial date. It has been known, however, that in the presence of obstacles (tube walls, wire screens, porous media matrix, solid particles) the initially formed deflagration may undergo slow but progressively increasing acceleration ending up as a detonation. It has been observed that the deflagration-to-detonation transition (DDT) is often preceded by the development of turbulence, generation of Mach waves and their coalescence to form shock waves. Yet whether these effects are indeed responsible for the predetonational flame acceleration (as has been often suggested) or merely prominent by-products of the latter has not been definitely ascertained and, after nearly 50 years of research, the issue continues to be a matter of debate. Recently in the course of studying flame propagation through an inert porous matrix, that effectively has many common features with adiabatic capillary, we came to realize that the predetonational flame acceleration (the core of DDT) has little to do with turbulence or Mach waves, totally suppressed by the system, but rather stems from the friction between the burning gas and the matrix. The transition is triggered by a localized explosion which in turn is conditioned by the spatial gradient of induction time in the friction induced extended preheat zone formed ahead of the advancing flame. The phenomenon, therefore, is physically quite similar to shockless initiation of detonation in non-uniformly preconditioned gas. In porous media or in adiabatic capillaries, the effect of friction is very strong, making the process highly dissipative and, thereby, rather benign both for analytical and numerical exploration. In contrast to this, for combustion in moderately wide smooth-walled pipes the friction is weak and acoustic effects are among the most prominent features of the system. Yet, as it turned out, this does not alter the main outcome of the previous study: the presence of mild friction still may cause the flame acceleration and transition to detonation (Figures 5,6). This time, however, the predetonational induction distance becomes rather long which, on the other hand, is quite in line with the experimental evidence. At sufficiently strong friction the normal near-Chapman-Jouguet detonation is found to undergo a jump-wise hysteresic transition to a low-speed quasi-detonation dominated by dissipative affects

(Figure 7).

The phenomenon is well-known experimentally but this is apparently the first successful attempt of its theoretical description. Due to its highly dissipative nature the quasi-detonation has many technical advantages over conventional unconfined detonation dominated by hydrodynamic nonlinearities. In quasi-detonation the latter play only a supporting role and for understanding the basic physics involved may well be ignored.

For all its rather simple mathematical structure the pertinent model appears to be rich enough to capture many typical and quite non-trivial features of unconfined detonation, such as oscillatory and spinning propagation modes (Figure 8). These studies are partially reported in the papers [3,5] (Sec. E).

C.3. Self-Fragmentation of Near-Limit Flames.

The burner stabilized laminar flames of low Lewis number premixtures are known to suffer diffusive instability resulting in the formation of a non-planar cellular structure. The latter is most prominent in weak premixtures sensitive to the radiative heat losses. In such systems, the cellular flame often breaks up into separate caplike fragments which sometimes close up on themselves to form seemingly stationary spherical structures called flameballs. To visualize the dynamics of their formation, Kagan and Sivashinsky (*Combust. Flame*, **108**, 220, 1997) recently conducted a direct numerical simulation of the pertinent reaction-diffusion system on a plane.

In accordance with earlier theoretical predictions, it has been found that the continuous cellular flame is rather robust and can propagate with heat loss rates greater than the maximum that extinguishes the planar flame. Yet, at sufficiently high heat losses the flame breaks up into separate near circular fragments moving through the premixture while leaving a significant portion of the fuel unconsumed. As the level of heat losses is increased the propagating flamelet sharply slows down forming an almost radially symmetric structure closely resembling the stationary spherical flame-ball described in the previous studies. Due to the logarithmic 'tail' the two-dimensional formulation does not admit the ideally motionless flamelets. Yet the model clearly indicates a noticeable tendency toward their formation near the extinction limit. In any event, the numerical study provided strong evidence that apart from the propagating cellular flames and the stationary flame-balls there is an intermediate combustion mode realized through localized yet traveling ball-like flamelets. The objective of the current study is to describe their structure and relation to the stationary flame-balls by means of an analytically tractable one-dimensional model. This study is reported in the paper [6] (Sec. E).

C.4. Stability of Counterflow Stabilized Flames.

The unrestricted planar premixed flame subject to a uniform stretch is known to be unconditionally stable. This is true for the flames predisposed to the diffusive as well as hydrodynamic instability. The reason for such an outcome is of purely kinematic origin. The stretch flattens out any flame disturbance by an unlimited enlargement of its length-scale while the resulting planar perturbations keep the flame stable. This, however, contradicts both the experimental and numerical observations showing that the flame destabilization is feasible provided the applied stretch is not too strong.

The present study shows that the origin of the above discrepancy is the system's overall-length-scale, d. The finiteness of the scale combined with invariably present boundary conditions serves as a permanent source of the d-scale disturbances. The latter, in turn, may well induce the instability, provided the flame stretch is weak enough.

The phenomenon is related to the formation of star-shaped and rotating polyhedral flames occurring in low-Lewis number premixtures and to the formation of multiple-cusp structures often observed in large-scale flames. The results obtained are reported in the paper [7] (Sec. E).

C.5. Chaotic Dynamics in Premixed Combustion.

Thermal feedback in exothermal chemical reactions can lead to a rich variety of non-relaxational dynamical behavior. One of the simplest systems of this kind is the so-called gasless combustion where solid fuel is transformed directly into solid product without intermediate gas phase formation.

Solid phase combustion is currently being studied as a new and potentially more effective means of synthesizing ceramic and metallic materials which have greater tolerances to high temperature and superior mechanical and electrical characteristics. Previous results, both theoretical and experimental, have indicated the various modes of propagation are possible. In addition to uniformly propagating planar fronts, planar fronts whose propagation velocities are oscillatory in time have also been observed. Under other conditions the reaction rate is not uniformly distributed on the sample cross-section but concentrated near its lateral surface in the form of a small luminous point moving in a helical fashion along the sample surface.

There are two physically related but mathematically distinct models to describe gasless combustion. Both involve a coupled set of equations for temperature and the limiting concentration of the fuel. In the first model the chemical reaction rate is governed by the distributed Arrhenius kinetics. Yet since the reaction rate is strongly temperature dependent the bulk of the chemical reaction occurs in a thin layer outside of which the reaction may be neglected. The reaction rate may thus be effectively replaced by a localized source whose position is determined by the overall dynamics of the system. One thus arrives at an essentially different, free-interface, model of the process in which the reaction rate is treated as a surface δ-function. The free-interface model is highly convenient for analytical studies. Contrary to this, numerical simulations of gasless combustion dealt until now exclusively with the reaction-diffusion formulation based on a distributed reaction rate. One of the fascinating findings unveiled in these numerical studies is the possibility of the irregular jumpwise advancement of the reaction zone, provided the system is shifted sufficiently deeply into the instability domain.

The natural question arising in this connection is whether such a complex dynamics is essentially due to the distributed nature of the reaction rate, or whether it can be covered by the free-interface formulation as well?

The numerical study undertaken in this paper shows that the δ-function based model is indeed rich enough to capture the main features of the above phenomena. The model exhibits a remarkable wealth of dynamical scenarios such as finite and infinite sequences of period doubling: the latter leading to chaotic oscillations, reversed sequences, Shilnikov type dynamics and infinite period bifurcations that replaces supercritical Hopf bifurcation. These studies are reported in the paper [8] (Sec. E).

C.6. The Effect of Stirring on the Limits of Thermal Explosion.

Since the early works of Semenov and Frank-Kamenetsky it has been known that insufficient heat transfer to the walls can lead an exothermically reacting stagnant premixture, e.g. enclosed between plates, to a fast-burning regime (explosion) whereas more intense transfer allows for a steady removal of the heat released in the reaction. The existence of an explosion threshold is due to exponential temperature dependence of the reaction rate. The process is thus inherently nonlinear.

Thermal explosions of the reactive mixtures are among the most fundamental phenomena of premixed combustion and, hence, one of the central topics of the field and its applications, e.g. to safety problems. It is, therefore, of considerable interest to elucidate how various complicating factors can modify the classical findings of Semenov and Frank-Kamenetsky.

For example, due to buoyancy, gradients of the temperature field may induce convective flows which, in turn, may affect the energy balance and thereby influence the explosion limits.

It is regarded as generally accepted that stirring, e.g. due to natural convection, promotes heat extraction from the system and thereby should raise the explosion threshold. Yet, as has been recently shown by Joulin & Sivashinsky (*Combust. Sci. Techn.* **77**, 329-335 (1991)), this conclusion may be wrong. Upon studying the interplay between natural convection and the explosion of the exothermically reacting fluid confined between two poorly conducting horizontal plates it was found that whereas short-scale convection does indeed hamper the explosion through the increased effective thermal diffusivity, the presence of large-scale eddies may have the opposite effect. The large-scale eddies distort the temperature field producing the so-called hot spots which may cause a local acceleration of the reaction thereby promoting the explosion.

In the above reaction-advection system the flow-field is not prescribed but results from the Rayleigh-Benard instability controlled by the external boundary conditions. Its evaluation is a difficult nonlinear problem that even in absence of heat release may be effectively performed only under specific limiting conditions, e.g. poor conductivity of the plates and weak heat release (Joulin & Sivashinsky, 1991). However, as has been mentioned in the latter study, the basic nature of the stirring promoted explosion does not depend on the origin of the convective eddies and the phenomenon is likely to manifest itself in a more straightforward approach where the background flow-field is regarded as prescribed rather than being one of the unknowns.

Adopting this approach the present paper addresses the conventional Frank-Kamenetsky model augmented with the advection term based on a prescribed periodic array of eddies. Analytical and numerical analyses reveal that, in accordance with the intuitive expectations, the short-scale stirring makes the thermal explosion more difficult through the increased heat transfer to the boundaries. However, under the long-scale stirring, promoting formation of hot spots, transition to the explosion may be facilitated rather than hampered.

This study is reported in the paper [9] (Sec. E).

D. Figures.

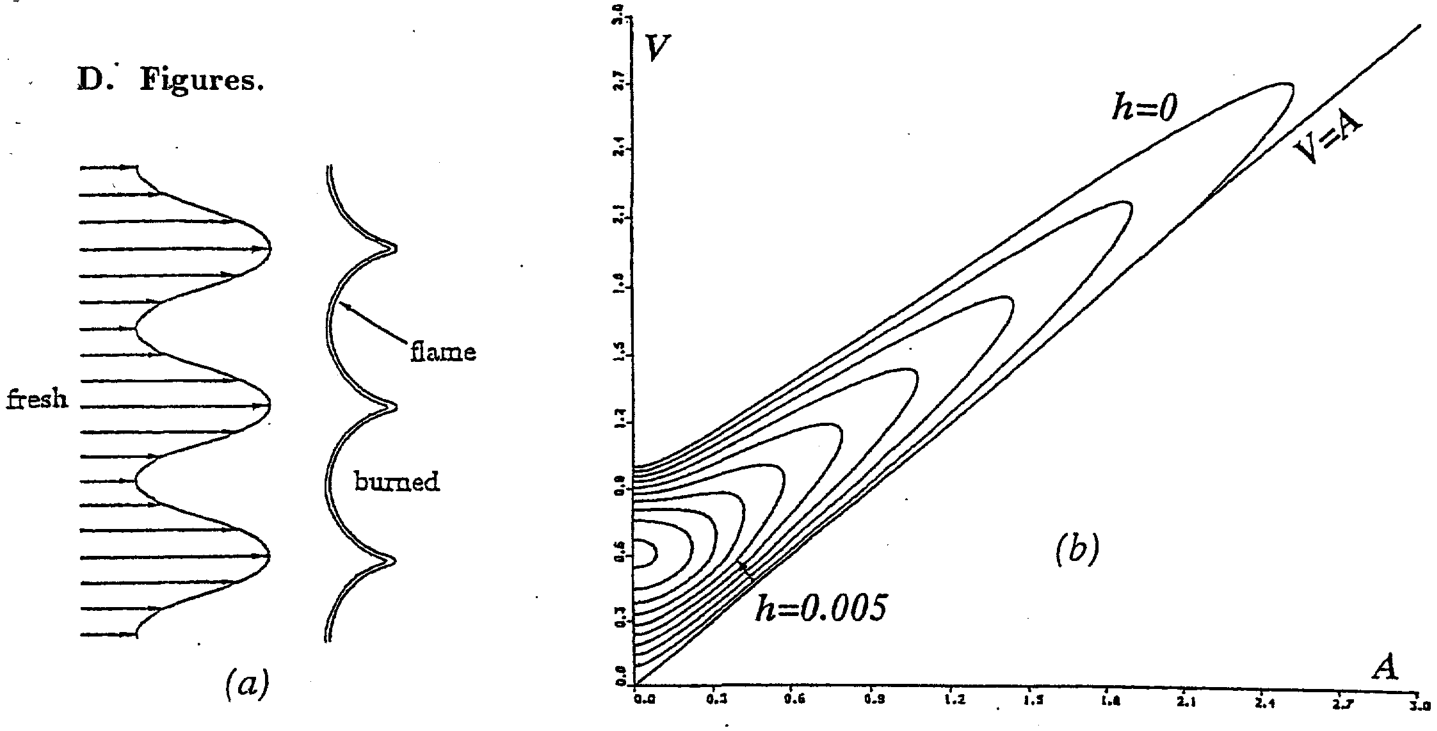


Figure 1. (a) - Diagram of the corrugated flame stabilized in periodic shear flow *u=V+A cos ky, v=o.* (*b*)- Flame speed (*V*) versus flow intensity (*A*) evaluated for different levels of heat loss (*h*) at *k=0.01* employing SVF analysis *(Brailovsky & Sivashinsky, Phys.Rev.51 (1995)1172-1183.*

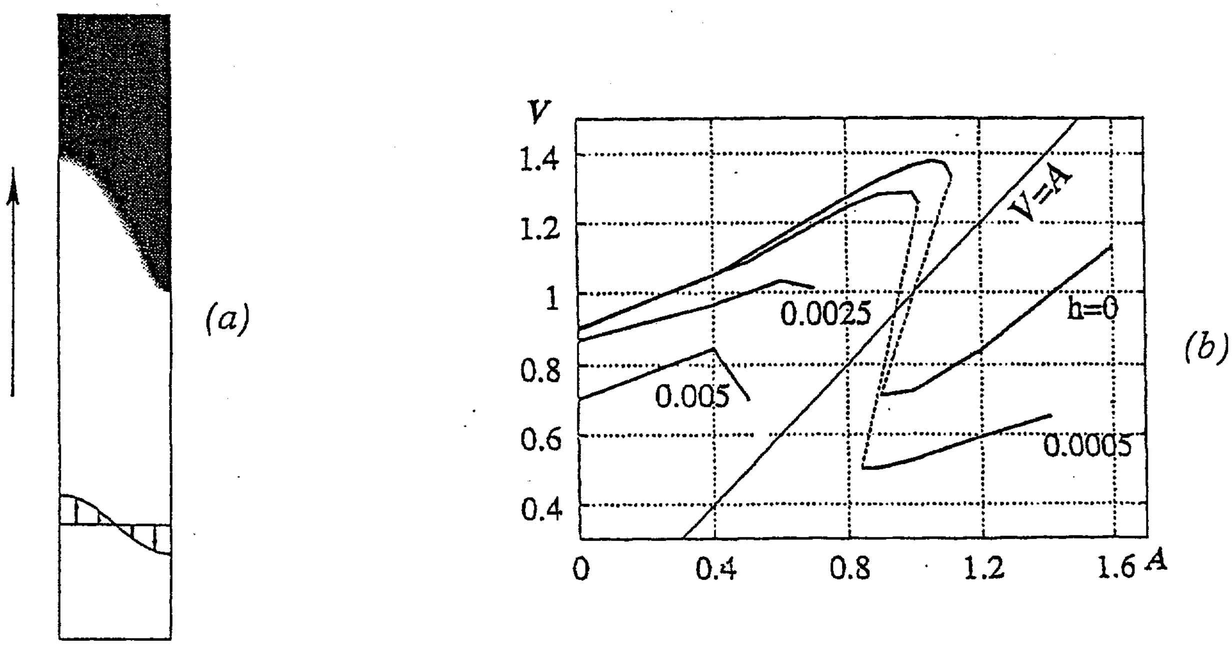
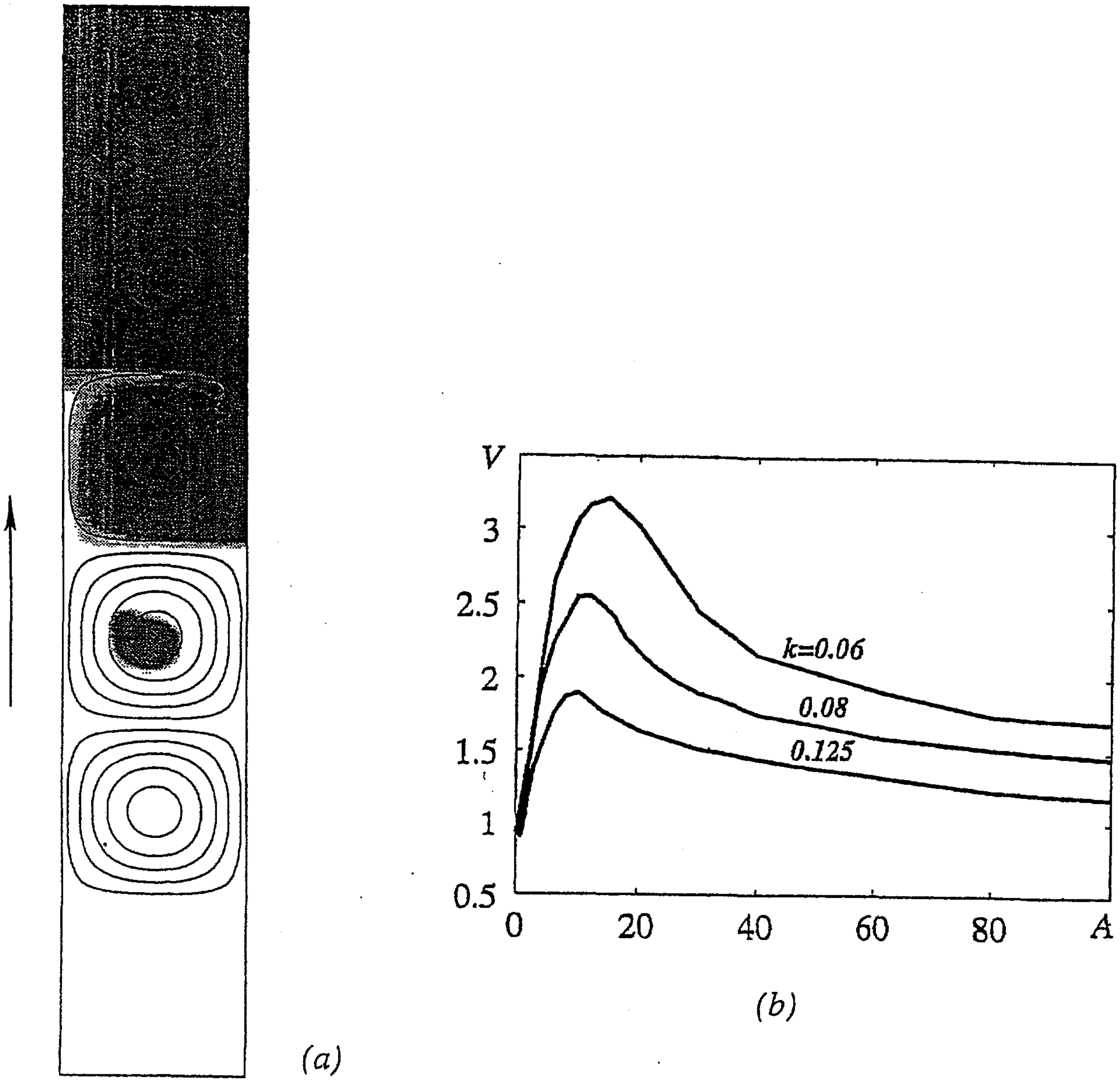


Figure 2. (a) - Distribution of the deficient reactant concentration (in premixed flame spreading through periodic shear flow *u=V÷A cos ky, v=o.* (b) - Flame speed (*V*) versus flow intensity (*A*) evaluated for different levels of heat loss (*h*). Numerical simulation of the pertinent reaction-diffusion-advection model for *k=0.06* and *h=0. 0.0005. 0.0025. 0.005.*



(a)

Figure 3. (a) - Distribution of the deficient reactant concentration (*C)* in premixed flame spreading through double-periodic flow $u = A \sin kx \cos ky$, $v = -A \cos kx \sin ky$, when the flame dynamics involves formation of the unburned gas pockets. (b) Overall flame speed (*V*) versus flow intensity (*A*) evaluated for different wave numbers (*k*). Numerical simulation of the pertinent reaction-diffusion-advection model at $h = 0$ and $k = 0.006, 0.08, 0.125$.

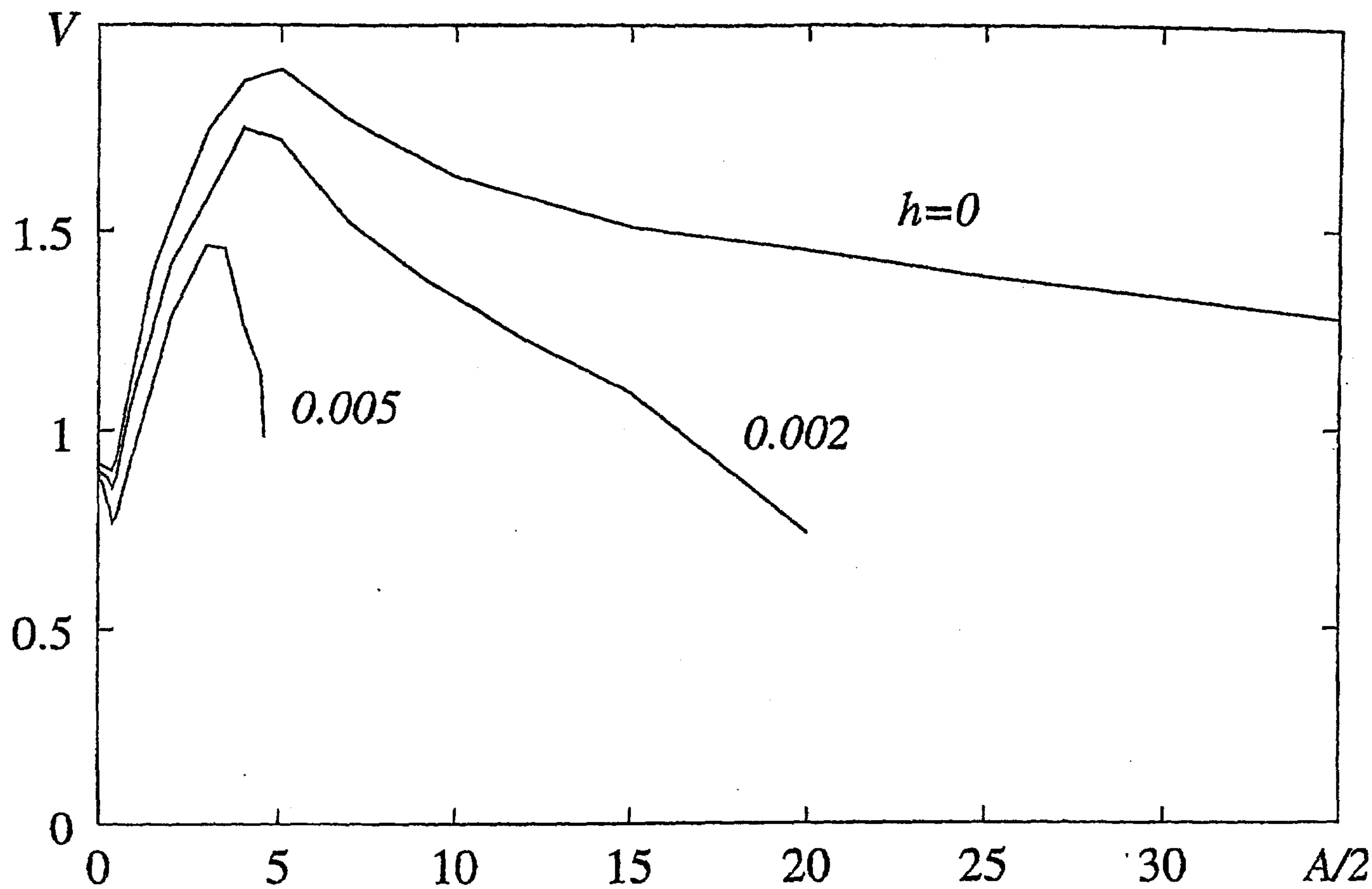


Figure 4. Overall flame speed (V) versus flow-field intensity (A) evaluated for different levels of heat loss (h). Numerical simulation of reaction-diffusion-advection model for the double-periodic flow $u = A \sin kx \cos ky$, $v = - A \cos kx \sin ky$ at $k = 0.125$, $h = 0$, 0.002, 0.005.

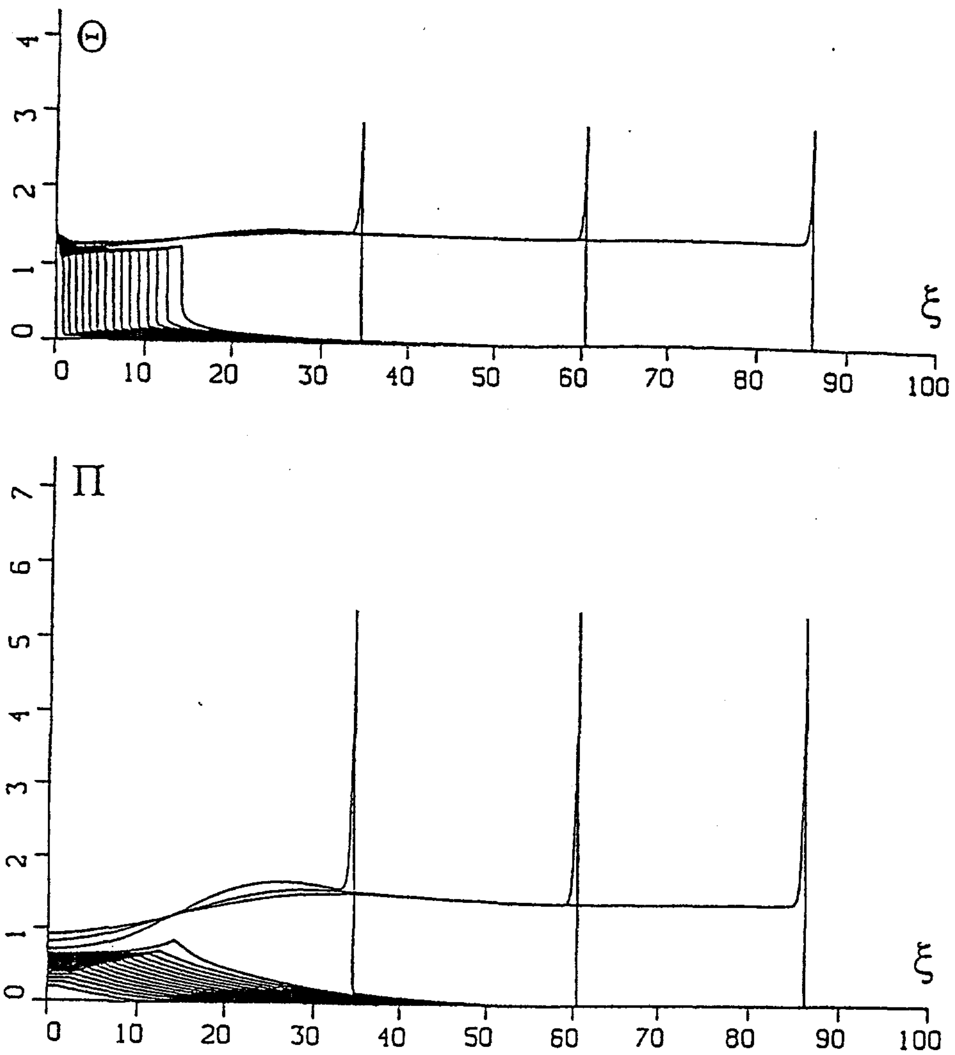


Figure 5. Profiles of pressure (Π) and temperature (Θ) at several equidistant instants of time.

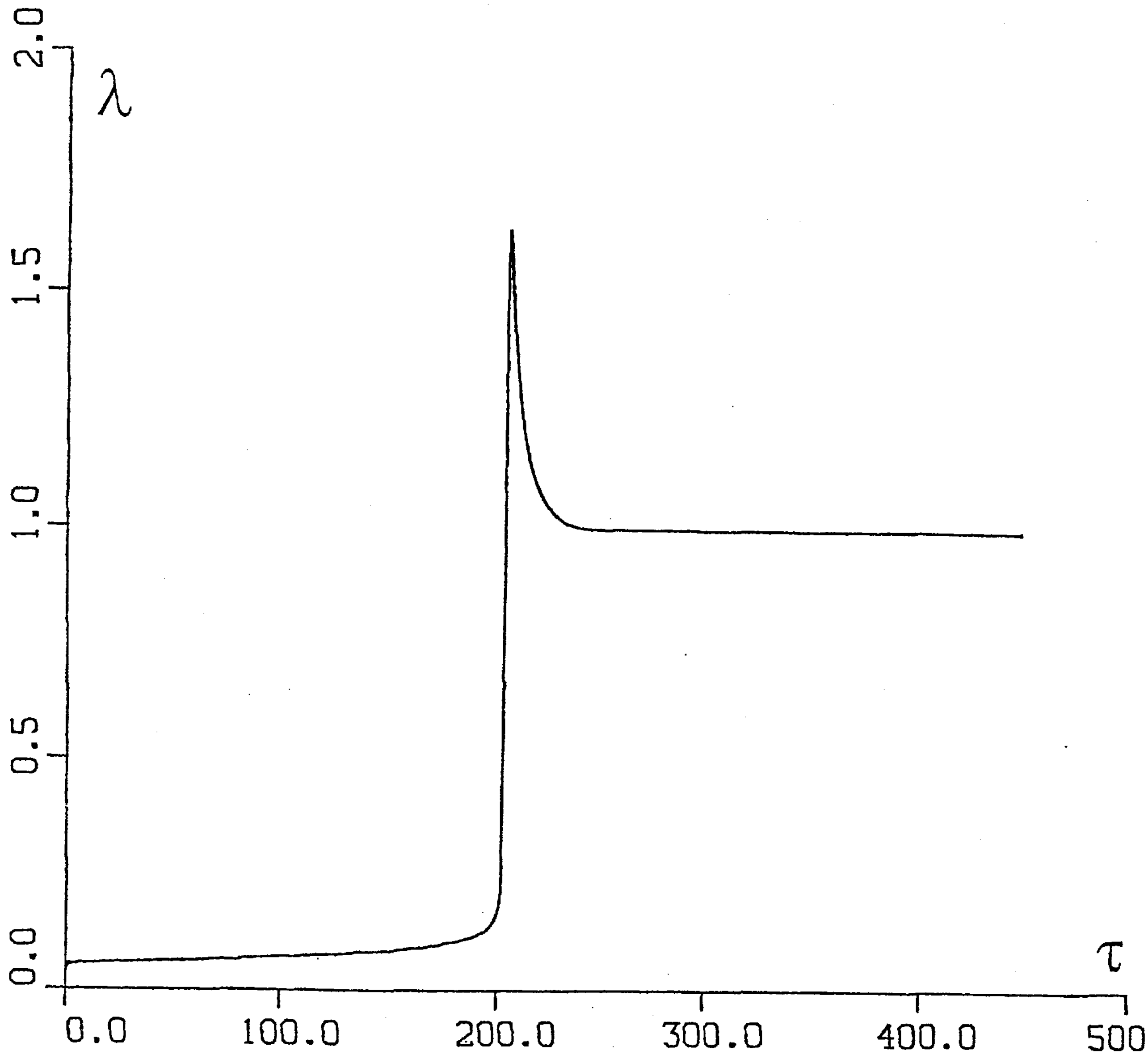


Figure 6. Temporal evolution of the reaction wave velocity (λ) adjacent to the point of deflagration-to-detonation transition.

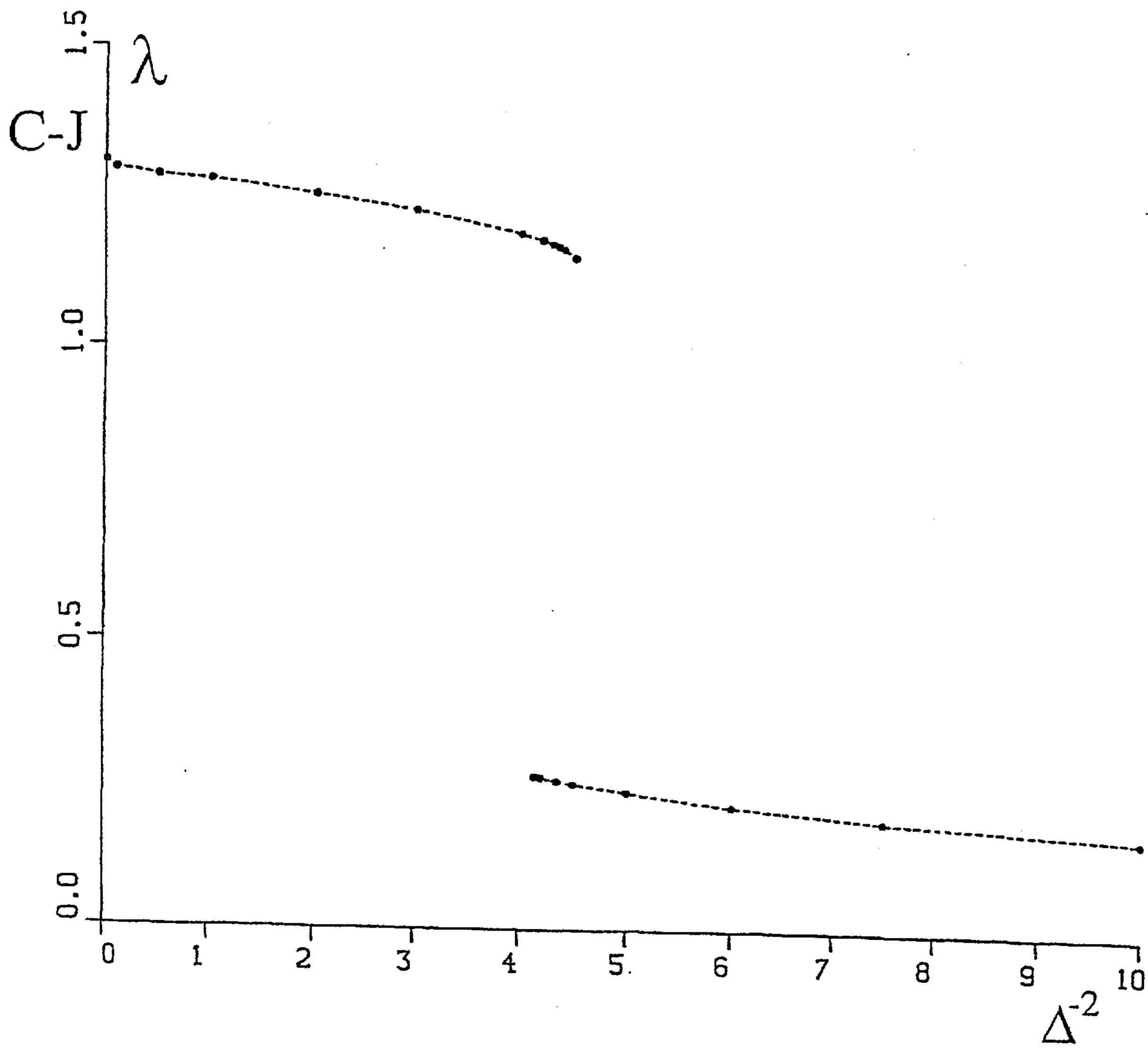


Figure 7. Propagation velocity of well settled detonation (λ) versus friction intensity (Δ^{-2})

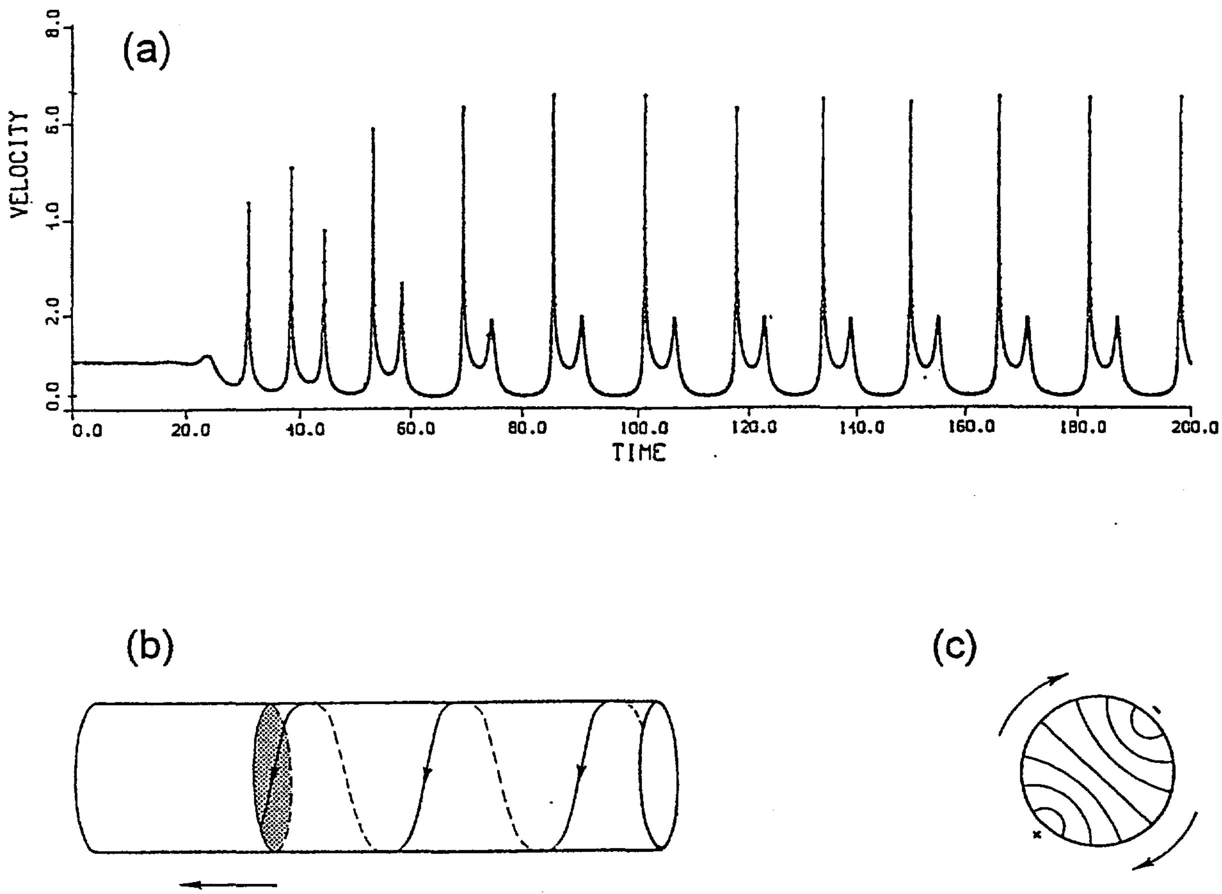


Figure 8. (a) - Oscillatory mode of quasi-detonation. Temporal evolution of the detonation wave velocity. (b) - Spinning mode of quasi-detonation. Diagram shows the trajectory of maximum temperature. Shaded cross-section corresponds to instantaneous position of the reaction wave front. Horizontal arrow indicates direction of wave propagation. (c) Distribution of isotherms over tube cross-section when detonation propagates in one-headed spinning regime. Plus and minus signs correspond to maximum and minimum temperatures, respectively. Arrows indicate one of the possible senses of rotation.

E. List of Resulting Publications.

1. L. Kagan, G.I. Sivashinsky, G. Makhviladze
   On flame extinction by a spatially periodic shear flow.
   Combustion Theory and Modelling 2, 399-404 (1998).
2. L. Kagan, G.I. Sivashinsky
   Flame Propagation and Extinction in Large-Scale Vortical Flows.
   Combustion and Flame 120, 222-232 (2000).
3. I. Brailovsky, V. Goldshtein, I. Shreiber, G.I. Sivashinsky
   On combustion waves driven by diffusion of pressure.
   Combustion Science and Technology 124, 145-166 (1997).
4. I. Brailovsky, G.I. Sivashinsky
   On deflagration-to-detonation transition.
   Combust. Sci. Techn. 130, 201-231 (1997).
5. I. Brailovsky, G.I. Sivashinsky
   Momentum loss as a mechanism for deflagration-to-detonation transition.
   Combustion Theory and Modelling 2, 429-447 (1998).
6. I. Brailovsky, G.I. Sivashinsky
   On stationary and travelling flame balls.
   Combustion and Flame 10, 524-529 (1997).
7. Y. Kortsars, I. Brailovsky, G.I. Sivashinsky
   On hydrodynamic instability of stretched flames.
   Combustion Science and Technology 123, 207-225 (1997).
8. M. Frankel, V. Roytburd, G.I. Sivashinsky
   Complex dynamics generated by a sharp interface model of self-propagating high-temperature synthesis.
   Combustion Theory and Modelling 2, 479-496 (1998).
9. L. Kagan, H. Berestycki, G. Joulin, G. Sivashinsky
   The effect of stirring on the limits of thermal explosion.
   Combustion Theory and Modelling 1, 97-111 (1997).