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CONTENTS

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1. INTRODUCTION

Considerable progress has recently been achieved in the understanding of the nature and character of spontaneous instabilities in premixed flames. The present survey is devoted to the latest theoretical results in this area, which have disclosed a deep affinity between flames and other nonequilibrium physical systems capable of generating regular and intrinsically chaotic structures.

1.1 Premixed flames. It is well known that the rate of a chemical reaction (W) in a gaseous mixture is an increasing function of temperature; usually $W \sim exp(-E/RT)$, where E is a constant, specific to the reaction and called its activation energy, and R is the universal gas constant. The larger E, the stronger the temperature-dependence of the reaction rate. Under normal conditions (atmospheric pressure, room temperature), the reaction rate in the majority of combustible mixtures is negligibly small. At sufficiently high temperatures, however, the reaction will take place at a substantial rate. When a combustible mixture is ignited at some point, e.g. by a spark, a rapid exothermic reaction is initiated; via conduction, this causes the temperature to rise in the adjacent layer of the mixture, inducing a chemical reaction there, and so on. Thus, the reaction, once begun, will spread through the mixture, converting it into combustion products. This self-sustaining wave of an exothermic reaction is known as a *premixed flame*.

The thermal mechanism of flame propagation just described has been known for a long time (Mallard & LeChatelier 1883), but it was not until the work of Zeldovich and

Frank-Kamenetsky (1938) that a really sound theory was formulated for the problem of steady plane flame propagation. Typical profiles of temperature (T), concentration (C), and reaction rate (W) in a one-dimensional combustion wave are shown in Fig. 1. T_u is the temperature of the unburned cold mixture, at which the reaction rate is negligibly small. C_u is the initial concentration of the reactant that is entirely consumed in the reaction (limiting reactant). T_b is the temperature of the burned gas, usually 5 to 10 times T_u. The thermal thickness l_{th} of the flame is defined as D_{th}/U_u, where D_{th} is the thermal diffusivity of the mixture and U_u the propagation speed of the flame relative to the unburned gas.

As the reaction rate is strongly temperature-dependent ($E/RT_b \sim 20$), the bulk of the chemical reaction occurs in a narrow temperature interval $\Delta T \sim RT_b^2/E$ around the maximum temperature T_b. This temperature region corresponds to a thin layer of width $\sim (RT_b/E)l_{th} = l_r$, outside which the chemical reaction may be neglected.

The propagation speed of the flame (U_u) is determined by balance between the quantity of heat liberated during the reaction and the heat required to preheat the fresh mixture:

$$U_u \sim \sqrt{D_{th} W(T_b)} \,. \tag{1}$$

For one of the most rapidly burning mixtures ($2H_2+O_2$), one has $U_u \simeq 10$ m/sec and $l_{th} \simeq 0.0005$ cm. For one of the most slowly burning mixtures (6% CH_4 + air), $U_u \simeq 5$ cm/sec and $l_{th} \simeq 0.05$ cm.

The mathematical theory of steady plane flames is now – in principle – complete, and there are several monographs on the subject (e.g., Williams 1965, Frank-Kamenetsky 1969, Kanury 1975, Glassman 1977, Zeldovich *et al.* 1980, Buckmaster & Ludford 1982). The situation is far less satisfactory in regard to non-steady phenomena in curved flames. In particular, until recently there was a considerable gap between theoretical and experimental results concerning the stability of a premixed flame front. The marked progress achieved in this field over the past five years is largely due to the asymptotic methods which have penetrated combustion theory from other, more classical areas of fluid mechanics.

1.2. Observed instabilities. For combustion to be actually maintained in the form of a steady plane reaction wave, the structure in question must be stable under small disturbances. Many flames are known to behave like one-dimensional reaction waves under normal laboratory conditions. However, some experiments have shown that there is a class of flames that prefer a characteristic two- or three-dimensional structure rather than a plane flame. It has long been known that the flame on a Bunsen burner may split up into triangular flamelets, which form – instead of the usual cone – a polyhedral pyramid, sometimes even rotating about its vertical axis (Smithells & Ingle 1882, Smith & Pickering 1928). Later it was shown (Zeldovich 1944, Markstein 1949) that this manifestation of instability is not unique. In combustion in wide tubes, the flame frequently breaks up into separate cells, ~1 cm in size, in a state of constant subdivision and recombination (Fig. 2). It was noticed that cellular structure tends to appear when the combustible mixture is deficient in the light reactant (e.g., rich hydrocarbon-air or lean hydrogen-air mixtures). Later it was pointed out that lean hydrocarbon-air mixtures are also not absolutely stable. Under the action of external large-scale disturbances, an originally smooth flame may exhibit sharp folds which are maintained under further deformation and extension of the flame (Fig. 3). Moreover, recent experiments on the propagation of large-scale flames in unconfined vapor clouds have shown that cellular instability may also appear in lean hydrocarbon-air mixtures, with cell size ~10 cm (Lind & Whitson 1977, Ivashchenko & Rumiantsev 1978). Finally, we should mention one of the most recent results, concerning spinning propagation of luminous flames (Gololobov *et al.* 1981). Thus, even freely propagating flames represent an extremely rich physical system.

1.3. Fundamental assumptions of the theory. A theoretical description of flame propagation requires simultaneous solution of an equation system including both the reaction-diffusion equations for each of the species and the equations of motion of the gaseous mixture. Since the propagation speed of the flame is significantly less than the speed of sound, effects due to the dynamic compressibility of the gas may be neglected. Hence, the density of the gas may be considered a function of temperature and concentrations only.

It turns out that the approximation representing the combustion reaction as a simple scheme:

$$\text{Fuel} + \text{Oxidant} \rightarrow \text{Products}$$

is sufficient for a qualitative description of the overwhelming majority of fluid-mechanical phenomena in premixed flames. It is convenient to consider the reactants as small additives to some "inert" gas; this justifies use of the independent-diffusion approximation in calculating their diffusion rates. Moreover, this assumption enables one to consider the gas almost homogeneous and thereby to simplify the equation of state. The "inert" gas (= dilutant) in experiments is frequently nitrogen.

As the reaction rate is strongly temperature-dependent ($E/RT_b \gg 1$), the reaction zone may be considered infinitesimally narrow in comparison with the thermal thickness of the flame ($l_r \ll l_{th}$). Thus, if one is interested in dimensions of the order of l_{th} or higher, it may be assumed that the reaction is concentrated on a certain surface – the flame front. The reaction rate (W) may therefore be replaced by a localized source, the intensity of which is determined by considering the processes that take place within the reaction zone. This problem may be solved in the spirit of Zeldovich and Frank-Kamenetsky (1938). For example, if the mixture is strongly non-stoichiometric (say, the concentration of the oxidant is much higher than that of the fuel), then the intensity of the localized reaction rate is proportional to $exp(-E/2RT_f)$, where T_f is the temperature on the curved, nonsteady flame front, which may differ from T_b by a quantity of the order of RT_b^2/E.

2. THERMO-DIFFUSIVE FLAME INSTABILITY

2.1. One-reactant linear theory. As early as 1944, Zeldovich proposed the following qualitative explanation of the observation that cellular flames tend to form in mixtures which are deficient in the light reactant. Consider a curved flame front. It is readily seen that conduction of heat tends to decrease the curvature of the flame, i.e., it is a stabilizing influence. Indeed, the parts of the chemical reaction zone which are convex toward the fresh mixture give out more heat than in a plane flame. The resulting cooling of the reaction zone slows down the forward-rushing parts of the flame. The concave parts, conversely, give out less heat than in the plane case, and so the temperature increases, and hence also the reaction rate. The concave parts of the flame move forward at a higher speed than in a plane flame. Thus the surface of the curved front is smoothed out.

Diffusion, however, has the opposite effect. The parts of the reaction zone convex toward the fresh mixture receive more fuel than in a plane flame. The reaction rate in the convex parts increases and the front curvature becomes greater. Thus diffusion has a destabilizing effect.

Hence it is clear that if the molecular diffusivity D_{mol} of the limiting reactant is sufficiently greater than the thermal diffusivity D_{th} of the mixture, one can expect a plane flame to be unstable. In the opposite case the flame front should be smooth.

The motion of a curved flame front is invariably accompanied by motion of the gas. However, it is evident from the foregoing qualitative arguments that hydrodynamic effects play a merely secondary role in the onset of thermo-diffusive instability. For a theoretical analysis of the phenomenon, therefore, it makes sense to disregard them. Formally, this may be done by assuming that the density of the gas is a constant. In that case thermal disturbances in the flame cannot be transformed into hydrodynamic disturbances, and so the problem of hydrodynamics is completely divorced from the problem of combustion proper. In other words, when studying the motion of the flame one can consider the hydrodynamic field to be assigned in advance.

In the model just described, linear analysis of the stability of a plane flame front to *long-wave* disturbances yields the following dispersion relation (Barenblatt *et al.* 1962):

$$\sigma = D_{th}[\tfrac{1}{2}\beta(1-Le)-1]k^2 \tag{2}$$

where $\beta = E(T_b - T_u)/RT_b$; $Le = D_{th}/D_{mol}$ is the Lewis number of the limiting reactant, which is assumed to be strongly deficient; σ is the rate of instability parameter; $\vec{k}$ is the wave vector of the disturbance of the flame front, $F \sim exp(\sigma t + i\vec{k}\cdot\vec{x})$. Thus, in agreement with the previous qualitative analysis, the flame is stable only if the mobility of the limiting reactant is sufficiently low ($Le > Le_c = 1-2/\beta$). At $Le < Le_c$ the flame is unstable. In a typical flame, $\beta \simeq 15$, and so $Le_c \simeq 0.87$.

However, as was pointed out later (Sivashinsky 1977a), a flame, though possibly unstable to long-wave disturbances, is nevertheless always stable to short-wave disturbances. At $Le \simeq Le_c$ the dispersion relation incorporating this relaxation effect of short-wave disturbances is

$$\sigma = D_{th}[\tfrac{1}{2}\beta(1-Le)-1]k^2 - 4D_{th}l_{th}^2k^4\,. \tag{3}$$

Thus, when the flame is unstable ($Le < Le_c$), there is a wavelength λ_c corresponding to the highest amplification rate of small disturbances (maximum σ).

2.2. Nonlinear theory. What happens to the flame front after the development of progressive disturbances?

First, it is readily seen that the dispersion relation (3) may be expressed as a linear equation for the disturbance of the flame front:

$$F_t + D_{th}[\tfrac{1}{2}\beta(1-Le)-1]\nabla^2 F + 4D_{th}l_{th}^2\nabla^4 F = 0 \tag{4}$$

This equation obviously yields exponential amplification of long-wave disturbances at $Le < Le_c$. In reality, this amplification will be checked by effects represented by certain nonlinear terms not present in Eq. (4). The structure of these terms may be established via the following semi-heuristic arguments.

Consider a curved flame front in the constant-density model. If the characteristic radius of curvature of the flame is significantly greater than its thermal thickness l_{th}, then the propagation speed of the flame relative to the gas may be considered a constant, equal to U_b. In a coordinate system at rest with respect to the undisturbed plane flame, the front $z = F(x,y,t)$ of such a curved flame is described by the eikonal equation:

$$F_t = U_b(1-\sqrt{1+(\nabla F)^2}) . \tag{5}$$

Near the stability threshold $Le \simeq Le_c$ one expects that $(\nabla F)^2 \ll 1$. Hence

$$F_t + \tfrac{1}{2} U_b(\nabla F)^2 = 0 . \tag{6}$$

Comparing this weakly nonlinear equation, which disregards effects due to distortion of the flame structure, with Eq. (4), in which these effects are included, one reaches the reasonable conclusion that $\tfrac{1}{2} U_b(\nabla F)^2$ is precisely the nonlinear term missing from Eq. (4). We thus obtain the following equation for the nonlinear evolution of the disturbed flame front:

$$F_t + \tfrac{1}{2} U_b(\nabla F)^2 + D_{th}[\tfrac{1}{2}\beta(1-Le)-1]\nabla^2 F + 4D_{th}l_{th}^2\nabla^4 F = 0 . \tag{7}$$

This equation is a rigorous asymptotic relation derived from the constant-density flame model, provided $Le - Le_c$ is a small parameter (Sivashinsky 1977b).

Thus the main nonlinear effect in the evolution of the disturbed flame front turns out to be of a purely kinematic nature. Qualitative arguments in favor of such a mechanism of nonlinear stabilization were put forward in the past by Manton *et al.* (1952), Markstein (1952), Petersen & Emmons (1961), Shchelkin (1965), and Zeldovich

(1966), whose point of departure was the Huygens principle. Eq. (7) may be put in a nondimensional, parameter-free form which is very convenient for numerical experimentation:

$$\Phi_\tau + \tfrac{1}{2}(\nabla\Phi)^2 + \nabla^2\Phi + 4\nabla^4\Phi = 0 . \qquad (8)$$

Numerical experiments on this equation have shown that when a plane flame is disturbed it ultimately evolves into a cellular flame, with characteristic cell size somewhat greater than λ_c. This structure was essentially nonsteady, the calls being in a state of continual *chaotic* self-motion (Michelson & Sivashinsky 1977) (Fig. 4). The chaotic behavior of cellular flames is indeed well known from the classical experiments of Markstein (1949,1964). This phenomenon was recently reconfirmed in experiments performed by Sabathier *et al.* (1981) under carefully controlled flow conditions, which prevented turbulence in the upstream flow (Figs. 2,3).

Thus, despite its simplicity, the one-reactant constant-density model proved sufficiently rich not only to provide an adequate description of the sensitivity of flame stability to the composition of the mixture and to predict the characteristic size of the cells, but also to describe their chaotic self-motion. Quite likely the model also describes polyhedral rotating Bunsen flames (Buckmaster 1982b) and the apparently similar phenomenon of traveling waves that sometimes appear in place of chaotically recombining cells (Markstein 1964, Sabathier *et al.* 1981).

However, the range of validity of the constant-density theory is limited. The point is that, according to the theory, the cell size should increase indefinitely as the Lewis number goes through its critical value Le_c. This is in clear contradiction to experimental observations, which indicate that the cell size at the stability threshold is finite (Markstein 1964,1970). As we shall show below (Sec. 3.4), near Le_c the effects due to the thermal expansion of the gas become significant, and this completely alters the nature of the instability.

2.3. Stability of nearly stoichiometric flames. The case discussed above corresponds to a strongly nonstoichiometric mixture, when the depletion of the excess reactant can be neglected and its concentration considered constant. The model was extended to nearly stoichiometric flames by Sivashinsky (1980) and Mitani and Joulin (1981), who showed that the main functional relationships of the one-reactant theory remain intact provided that *Le* is interpreted as a suitably weighted average of the Lewis numbers of the fuel and the oxidant. This yielded an explanation of the interesting observation that cellular instability is observed in nearly stoichiometric mixtures even when there is a certain excess of the light component (Bregeon *et al.* 1978). A recently published paper of Mitani and Williams (1980) on nearly stoichiometric hydrogen-air cellular flames revealed that the predictions of the theory are also in qualitative agreement with the experimental data on cell size.

2.4. Effects due to acceleration. Since combustion is accompanied by thermal expansion of the gas, it is clear that buoyancy will exert a stabilizing effect on downward-propagating flames. As in the Boussinesq theory of natural convection, this effect may be incorporated through a *force* term, while remaining within the limits of the constant-density model (Matkowsky & Sivashinsky 1979). As a result, the dispersion relation (3) is augmented by addition of a stabilizing term $g(1-\varepsilon)/2U_b$, where $\varepsilon = \rho_b/\rho_u$ is the thermal expansion coefficient of the gas. The nonlinear equation (8) is modified to become

$$\Phi_\tau + \tfrac{1}{2}(\nabla\Phi)^2 + \nabla^2\Phi + 4\nabla^4\Phi + G\Phi = 0\,. \qquad (9)$$

Here G is a nondimensional parameter proportional to the reciprocal of the Froud number. When $G > 1/16$ combustion is stable; this should be the case in sufficiently slow flames (Markstein & Somers 1953). Near the stability threshold, chaotic fluctuations disappear and the flame takes on a steady, almost regular, cellular structure with cell size λ_c (Fig. 5).

2.5. Effects due to heat loss. It is known that in rich hydrocarbon-air mixtures cellular stability is likely to be observed near the flame propagation limit. Thus, one might expect that inclusion of heat loss should expand the range of unstable Lewis numbers. Analysis of flame stability in the case of volumetric heat loss has fully corroborated this conjecture (Joulin & Clavin 1979, Sivashinsky & Matkowsky 1981). With heat loss included, the dispersion relation (3) becomes

$$\sigma = D_{th}\left[\frac{\frac{1}{2}\beta(1-Le)-1-2ln\gamma}{1+2ln\gamma}\right]k^2 - D_{th}l_{th}^2\left[\frac{4+2ln\gamma}{\gamma^2(1+2ln\gamma)}\right]k^4 \tag{10}$$

where γ is the ratio of the propagation speed of the nonadiabatic flame to that of the adiabatic flame (U_b). As one approaches the flame propagation limit ($\gamma \rightarrow 1/\sqrt{e}$), Le_c tends to unity, i.e., the instability region expands. Simultaneously one observes a sharp increase in the instability rate σ. Thus, for example, a downward-propagating adiabatically stable flame may become unstable when heat loss is taken into account.

2.6. Effects due to stretching. In practical situations the flame is frequently situated in a nonuniform flow field and is therefore subjected to large-scale flame stretch (Karlovitz *et al.* 1953). Recently, Law *et al.* (1981), Ishizuka *et al.* (1982), and Ishizuka and Law (1982) have systematically investigated the extinction and stability limits of propane-air flames in stagnation-point flow, which imposes a well-characterized strain rate on the bulk flame. Their results show that, while lean flames are absolutely stable, in rich mixtures flame-front instability of various configurations may appear, depending on the strain rate. When the flow rate is so slow that the flame is situated in the nearly one-dimensional flow field close to the burner surface, the instability is exhibited in the form of a cellular flame. If one increases the flow rate, and thereby the stretch, the flame recedes from the burner surface and moves further into the stagnation flow. It becomes star-shaped, with diametrically oriented ridges. Finally, with sufficiently strong blowing, all instabilities are suppressed, and the flame becomes smooth. Thus stagnation-point flow stabilizes the flame.

The mathematical problem of stability turns out to be quite unusual, in that the classical stability analysis appears somewhat misleading. For example, in the two-dimensional version of the problem the stagnation-point flow is $\vec{v} = (-qx, qy)$, where q is the flow rate parameter. The one-dimensional version of Eq. (8) for the disturbance of a plane flame is modified to become (Sivashinsky *et al.* 1982)

$$\Phi_\tau + \tfrac{1}{2}\Phi_\eta^2 + \Phi_{\eta\eta} + 4\Phi_{\eta\eta\eta\eta} + \alpha(\eta\Phi)_\eta = 0 \tag{11}$$

where α is a nondimensional parameter proportional to q.

The solution of the linearized equation (11) may be expressed as a combination of functions of the type

$$\Phi = A(\tau) exp\,(ik\eta e^{-\alpha\tau})$$

where (12)

$$A_\tau = (k^2 e^{-2\alpha\tau} - 4k^4 e^{-4\alpha\tau} - \alpha)A\ .$$

In the initial instant of time there are growing modes if $\alpha < 1/16$. However, as $\tau \to \infty$ the amplitude of the disturbance vanishes for any positive α. This is because with elapsing time any harmonic disturbance is stretched, while a disturbance of infinitely long wavelength ($k = 0$) is damped out as $exp\,(-\alpha\tau)$. Thus the flame front would seem to be absolutely stable for any positive α. However, this does not agree with the cellular flame structure observed at small flow rates (i.e., small α).

The apparent contradiction is resolved by observing that the neglected nonlinear term represents mode interaction, which continually generates short-wave disturbances whose amplitudes may increase during certain time intervals. Hence, within the framework of the original nonlinear theory, one can expect a permanently excited state of the flame front if α is sufficiently small. Numerical experiments with the nonlinear equation (11) have confirmed that the flame front is cellularly unstable at $\alpha \le 0.01$ and stable at $\alpha \ge 0.02$.

The nonlinear instability mechanism just described also occurs, in the case of an expanding spherical or cylindrical flame. In the latter case, for example, the evolution equation is (Sivashinsky 1979):

$$\Psi_\tau + \frac{1}{2\tau^2}\Psi_\vartheta^2 + \frac{1}{\tau^2}\Psi_{\vartheta\vartheta} + \frac{4}{\tau^4}\Psi_{\vartheta\vartheta\vartheta\vartheta} = 0 \tag{13}$$

where Ψ is the disturbance of the expanding cylindrical front and ϑ is a suitably scaled polar angle. Normalizing Ψ by the growing flame radius ($\Psi = \tau\Phi$; Istratov & Librovich 1969) and introducing cartesian coordinates on the flame surface ($\tau\vartheta = \eta$), one can bring Eq. (13) to the following form:

$$\Phi_\tau + \tfrac{1}{2}\Phi_\eta^2 + \Phi_{\eta\eta} + 4\Phi_{\eta\eta\eta\eta} + \frac{1}{\tau}(\eta\Phi)_\eta = 0 . \tag{14}$$

The last term in this expansion can be interpreted as a stretch generated by the expanding flame. Because of this stretch, linear analysis implies that the flame is absolutely stable; hence it is inadequate. The difficulty may obviously be resolved by a device similar to that employed for the stagnation-point flame.

To conclude this section, we note that if *Le* is varied continuously with the flame stretch held fixed, the cell size remains finite at the stability threshold.

2.7. Oscillatory and spinning flames. Analysis of the full dispersion relation for the constant-density flame model shows that if $\frac{1}{2}\beta(Le-1) > 5$ the real part of the instability rate σ is positive, and so the flame may be unstable also at fairly large Lewis numbers (Sivashinsky 1977a). Since the imaginary part of σ does not vanish when the stability threshold is crossed, the new propagation mode induced by instability of a plane flame may consist of oscillations, traveling waves or even spinning.

In real adiabatic gaseous flames, the instability region is not likely to be reached. However, as shown by Joulin and Clavin (1979), if there is volumetric heat loss the instability region is considerably expanded, and a real flame may well become oscillatorily unstable. The first announcement of spinning flame propagation was made recently by Gololobov *et al.* (1981) in connection with an investigation of the acetylene decomposition flames. Since such combustion is accompanied by high radiation heat loss (due to intensive sooting), the effect observed may well be the first corroboration of the theoretical possibility discussed above. Earlier, oscillatory and spinning flames had been observed only in gasless combustion of condensed systems (Merzhanov *et al.* 1973). In these systems, the diffusivity of the fuel is zero ($Le = \infty$) and the instability region is easily reached. The pertinent theoretical analysis was presented in Matkowsky and Sivashinsky (1978) and in Sivashinsky (1981).

Qualitatively new modes of oscillatory instability were discovered in investigation of flames stabilized on flat porous flame-holders. A steady theory of such burners was developed by Carrier and Fendell (1978), Ferguson and Keck (1979), and Clarke and McIntosh (1980). It was observed (Margolis 1980,1981; Matkowsky & Olagunju 1981; Buckmaster 1982) that conductive heat loss to the burner may be sufficient to bring on oscillatory instability even for relatively small Lewis numbers. As shown recently by Joulin (1981,1982), flame oscillation is described by a delayed nonlinear differential equation of Hutchinson type:

$$\Phi_\tau + \tfrac{1}{2} B\{1 - exp[-\Phi(\tau-2)]\} = 0 \qquad (15)$$

where Φ is the displacement of the flame front relative to its equilibrium position, and B is a number defined by the physico-chemical parameters of the system.

If $B > \pi/2$, the equilibrium state ($\Phi = 0$) becomes unstable and the system begins to perform saw-tooth oscillations. This oscillatory mode is in agreement with earlier numerical studies and certain experimental observations (Margolis 1980, 1981).

3. HYDRODYNAMIC FLAME INSTABILITY

3.1. Linear theory. In our discussion of thermo-diffusive flame instability, we ignored the effect of thermal expansion of the gas and, by the same token, the interaction of the flame with the hydrodynamic disturbances that it generates. It was Darrieus (1938) and Landau (1944) who made the first analysis of flame stability, assuming that the flame is a density jump propagating at a *constant* speed in an incompressible, nonviscous, nonconducting fluid. This approach is quite legitimate if one is interested in disturbances of wavelength that considerably exceed the thermal thickness l_{th} of the flame. Since the Darrieus-Landau model does not include any characteristic length, it is clear that the instability rate σ must depend on the flame speed U_b and the disturbance wavevector $\vec{k}$ as follows:

$$\sigma = \Omega_0 U_b k \quad , \quad k = |\vec{k}| \tag{16}$$

where Ω_0 is a nondimensional function of the parameter $\varepsilon = \rho_b / \rho_u$ – the ratio of densities of the burned and unburned gas. $\Omega_0(\varepsilon) = (\sqrt{\varepsilon+\varepsilon^2-\varepsilon^3} - \varepsilon)/(1+\varepsilon)$ is positive, for all $\varepsilon < 1$, that is, the flame is unstable to disturbances of all wavelengths. However, this is in conflict with experiment. Under normal laboratory conditions, one often observes smooth steady flames stabilized in quite wide tubes (diameter ~15 cm).

According to Eq. (16), short-wave disturbances should increase at a higher rate than long-wave disturbances. But it is precisely for short-wave disturbances that the Darrieus-Landau model breaks down, since they induce distortion of the flame front structure and are liable to alter its propagation speed. For this reason, later work on hydrodynamic flame instability was aimed at correcting the Darrieus-Landau solution (16) in the region of short-wave disturbances. The first important work in this direction was due to Markstein (1951). Markstein suggested that one characterize the effect of the distortions of flame structure by a certain phenomenological constant, having the dimension of length, relating the flame propagation speed to the curvature of the front. The result was that a plane flame is stable to short-wave disturbances and unstable to long-wave disturbances. This would mean that a plane flame may be observed only when combustion takes place between walls that prevent the appearance of long-wave disturbances.

Markstein's model was then generalized by Eckhaus (1961), who showed that the speed of a curved flame also depends on the gradient of the tangential component of the gas velocity along the front (i.e., the flame stretch).

The Markstein theory yields stabilization of short-wave disturbances only when the phenomenological constant has a certain sign. However, it may also have the opposite sign, due, say, to the possibility of thermo-diffusive instability. In that case short-wave disturbances would provide an additional destabilizing factor.

The uncertainties inherent in the Markstein theory stimulated new research, in which flame instability was investigated taking the flame structure into consideration, i.e., effects due to heat conduction, diffusion, viscosity, and chemical kinetics. However, the difficulties encountered in the process were so great that the investigators were forced to make various arbitrary assumptions concerning the structure of the disturbed flame front, in order to avoid insurmountable mathematical problems. As a consequence, the results obtained were in conflict not only quantitatively but even qualitatively. For example, until recently it was unclear whether viscosity is a stabilizing or destabilizing factor in flames (Markstein 1964).

Istratov and Librovich (1966) were the first to realize that the determination of corrections to the Darrieus-Landau solution is a singular perturbation problem. The point is that long-wave disturbances of the flame front create hydrodynamic disturbances, extending on both sides of the front for a distance of the same order as the wavelength of the disturbance ($2\pi/k$). Thus, the structure of the disturbed flame has at least two characteristic lengths, l_{th} and $2\pi/k$ ($\gg l_{th}$). As there was then no systematic technique for the solution of such problems, a mathematically consistent implementation of this idea was achieved only recently by Frankel and Sivashinsky

(1982) and Pelce and Clavin (1982). They obtained the following two-term expansion of the rate of instability parameter σ:

$$\sigma = \Omega_0 U_b k - \Omega_1 D_{th} k^2 \tag{17}$$

where

$$\Omega_1 = \frac{\varepsilon(1-\varepsilon)^2 - \varepsilon \ln\varepsilon(2\Omega_0+1+\varepsilon)}{2(1-\varepsilon)[\varepsilon+(1+\varepsilon)\Omega_0]} - \frac{\varepsilon(1+\Omega_0)(\varepsilon+\Omega_0)\beta(1-Le)}{2(1-\varepsilon)[\varepsilon+(1+\varepsilon)\Omega_0]} \int_0^{1-1/\varepsilon} \frac{ln(1+\xi)d\xi}{\xi}.$$

The correction to the Darrieus-Landau solution turned out to be independent of the Prandtl number. Thus, unlike the effects of heat conduction and diffusion, viscosity exerts a secondary effect on flame stability, which manifests itself, apparently, in higher-order terms of the expansion in powers of kl_{th}. These terms may, however, be significant when $\Omega_1 < 0$, when hydrodynamic flame instability combines with thermo-diffusive instability, and also if Ω_1, when positive, is also small.

It should be noted that when $\varepsilon = 0.2$, Ω_1 is negative if $\beta(1-Le) > 2.66$. Thus, in comparison with the constant-density model, inclusion of thermal expansion of the gas somewhat narrows down the thermo-diffusive instability region.

When $\Omega_1 > 0$, Eq. (17) implies the existence of a wavelength λ_c corresponding to maximum amplification rate of small disturbances. If one assumes that the nonlinear evolution ultimately produces a structure with this characteristic length (Rayleigh principle) then, for example, for a typical slow flame ($\varepsilon = 0.2$, $\beta = 15$, $Le = 1.2$, $l_{th} = 0.2$ mm), one obtains $\lambda_c \simeq 100\, l_{th} \simeq 2$ cm). Structures with this characteristic cell size should have been observed in the combustion of lean hydrocarbon-air mixtures in wide tubes (~ 10 cm). However, as is well known, this is not the case. The nature of the onset of hydrodynamic instability is clarified only after nonlinear effects have been incorporated.

3.2. Nonlinear theory. Arguments analogous to those employed in the theory of thermo-diffusive instability (see Sec. 2.2) yield the following evolution equation for the flame front:

$$F_t + \tfrac{1}{2} U_b (\nabla F)^2 = \Omega_1 D_{th} \nabla^2 F + \Omega_0 U_b I\{F\}\,, \tag{18}$$

where

$$I\{F\} = \frac{1}{4\pi^2} \int_{-\infty}^{\infty} |\vec{k}| e^{i\vec{k}(\vec{x}-\vec{x}')} F(\vec{x}',t) d\vec{k} d\vec{x}'\,.$$

When the thermal expansion of the gas is low (i.e., weak hydrodynamic instability), Eq. (18) is a rigorous asymptotic expansion, which can be derived from the full hydrodynamic equation system of the flame (Sivashinsky 1977b).[3]

Contrary to the predictions of the linear theory, numerical experiments on the one-dimensional version of Eq. (18) in an interval of width 10 λ_c have shown that, during nonlinear evolution, there appear on the flame front surface only one or two steady folds, strongly pointed toward the burned gas (Michelson & Sivashinsky 1977). If the folds lie at the ends of the interval, the flame will obviously present a smooth surface, convex toward the fresh mixture. Thus, hydrodynamic instability alone is sufficient to ensure that a flame in a wide tube will be curved (Uberoi 1959). A curved flame generates a gradient in the tangential component of the gas velocity along the front (i.e., stretch), and this is apparently what gives this configuration its remarkable stability (Zeldovich *et al.* 1980,1981, see also Sec. 2.6).

Figure 6 shows the results of numerical solution of Eq. (18) in a square $5\lambda_c \times 5\lambda_c$. Such folds are frequently formed when the flame crosses various kinds of obstacles, such as electrodes or a widely-spaced wire grid (Markstein 1964, Palm-Leis & Strehlow 1969) (Fig. 3). In view of the stability of these folds (which are maintained even when the flame is stretched), it was suspected in the past that they are yet another variety

[3] In the limiting case of low thermal expansion, the gas flow is irrotational both ahead of the flame front and *behind* it. Thus, contrary to a periodically expressed opinion, the effect of vorticity generation on flame stability is not decisive.

of spontaneous flame instability (Markstein 1964, 1970).

The effects of diffusion and heat conduction are obviously most significant in the zone of the cusp, where the curvature of the front is extremely large. Outside this region the flame may be described perfectly well by the following truncated equation, corresponding to the Darrieus-Landau model:

$$F_t + \tfrac{1}{2} U_b (\nabla F)^2 = \Omega_0 U_b I\{F\} . \tag{19}$$

Since the Darrieus-Landau model does not involve any characteristic length, Eq. (19) permits the existence of flames in which the distances between consecutive folds are arbitrarily large. A recent analysis of this equation by McConnaughey (1982) has shown that ∇F has a logarithmic singularity at the cusps of the folds. Thus, the flame front is infinitely sharp at the cusps and, consequently, the structure of such flames is essentially different from that of a Bunsen wedge.

3.3. Thermal expansion induced cellular flames. If the distance between the folds is increased indefinitely, the stabilizing effects of stretching and curvature are weakened and one expects new folds to appear. Thus, in order to investigate the fully developed hydrodynamic instability one must consider sufficiently large-scale flames. Experiments of this type were recently carried out, in connection with the investigation of accidental industrial explosions, by Lind and Whitson (1977) and Ivashchenko and Rumiantsev (1978). Lind and Whitson experimented with lean hydrocarbon-air mixtures in 0.05 mm thick polyethylene film hemispheres of 5 m and 10 m radius. Ivashchenko and Rumiantsev carried out similar experiments in 0.05 to 0.08 mm thick rubber spherical shells of 2.5 m radius. It was noticed that, as the flame expanded, it became rough, with a "pebbled" appearance. This structure increased in size to about 0.4 to 1.0 m, with *finer* structure superimposed. It was observed that for systems with markedly different burning velocities the measured space velocity was 1.6 to 1.8 times the expected value, calculated from the normal burning velocity measured in the laboratory (Lind & Whitson 1977).

Ivashchenko and Rumiantsev also noted that when the sphere radius reached ~5 cm the flame became cellular, with cells about 1 to 2 cm in size. As the flame sphere grew the cell size increased, reaching ~6 to 10 cm for sphere radius ~0.3 to 0.5 m. The maximum speed was 1.5 to 2 times the speed of the undisturbed spherical flame.

Stimulated by these experimental observations, Michelson and Sivashinsky (1982) undertook new numerical experiments on Eq. (18), considering a wider interval (40 λ_c). They found that, alongside the deep folds on the flame, there indeed appeared a fine structure with a well-defined cell size (~5 λ_c). In the example cited previously, this implies a cell size of ~10 cm. Here the deep folds may evidently be associated with the large-scale structures observed by Lind and Whitson. The numerical experiments also indicate that the fine structure is nonsteady. The cells continually and chaotically recombine, as occurs in thermo-diffusive instability.

Of the earlier experimental observations of hydrodynamic instability in flames, we would like to mention the work of Simon and Wong (1953), studying flames in a rich methane-air mixture filling a soap bubble of initial radius ~5 cm. When the flame radius was ~1.5 cm, the initially smooth flame front took on a cellular appearance, and simultaneously the flame was seen to accelerate. However, the relatively small volume of the mixture did not permit a sufficiently developed cellular instability and the wrinkled flame did not reach the uniform propagation mode.

3.4. Cellular flames near the thermo-diffusive instability threshold. If Ω_1 is small (i.e., Le is near Le_c), higher-order terms of the expansion in powers of kl_{th} become important. This parameter range merits close attention, since it is here that the thermo-diffusive model of a cellular flame breaks down (see Sec. 2.2). Moreover, almost any mixture may be brought to this range if the ratio of the reactant concentrations is suitably adjusted. However, computation of higher-order terms for the dispersion relation (17) is an extremely cumbersome task. Up to the present, this has been done only in the case of low thermal expansion, when one actually obtains a linear combination of the dispersion relation (16) and the relation (3) of the thermo-diffusive theory (Sivashinsky 1977b). Numerical experiments on the corresponding fourth-order integrodifferential equation (Michelson & Sivashinsky 1977) have shown that, if $\Omega_1 = 0$ (i.e., $Le = Le_c$), one obtains a chaotically recombining cellular structure with *finite* cell size of the order of λ_c. The cells, remaining finite, disappear for small positive Ω_1, i.e., when Le becomes greater than Le_c. Thus, near Le_c the dominating factor responsible for cellular instability may be of a purely hydrodynamic nature. We emphasize that, regardless of whether Ω_1 is positive, the characteristic cell size in this case, as in the case of purely thermo-diffusive instability, is determined by the fourth-order terms. Moreover, since $Le_c < 1$, the temperature variation along the front is quite similar here to that in purely thermo-diffusive cells (Sec. 2). This is one of the essential differences between near-Le_c thermal-expansion induced cells and the large-scale cells discussed in Sec. 3.3.

3.5. Effects due to acceleration. Hydrodynamic instability of downward-propagating flames becomes weaker and may even be completely suppressed by buoyancy effects (see also Sec. 2.4). These effects were discussed in detail by Markstein (1964) in his phenomenological theory. The problem was recently reconsidered by Pelce and Clavin (1982), within the framework of a complete hydrodynamic flame model, consistently incorporating the effects of transport and chemical kinetics. In contradistinction to a freely propagating flame, a flame propagating in the presence of a stabilizing acceleration is stable to long-wave disturbances. Near the stability threshold, the unstable modes are concentrated near λ_c -- the wavelength corresponding to maximum amplification rate of σ. Consequently, hydrodynamic instability should here be manifested as cells of size $\sim\lambda_c$. This structure has indeed been observed in flames situated in a periodically varying acceleration field induced by vibrations of the gas flow (Markstein 1964,1970). We emphasize that cells of size ~1 cm appear here even in flames in lean hydrocarbon-air mixtures, which do not exhibit cellular instability under normal conditions.

A similar type of fixed-size structure may also be induced by oscillations of the flame-holder (Petersen & Emmons 1961) or by large-scale fluctuations of turbulent gas flow (Palm-Leis & Strehlow 1969).

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FIGURE CAPTIONS

Fig. 1. Profiles of temperature, concentration, and reaction rate in a one-dimensional combustion wave.

Fig. 2. Rich propane-air cellular flame in state of chaotic self-motion. (Courtesy of P. Clavin, University of Provence, Marseille. Originally in F. Sabathier *et al.* (1981).)

Fig. 3. Free flame balls obtained in a low-speed laminar flow by spark ignition. (a) Cellularly stable lean butane-air flame. (b) Cellularly unstable lean hydrogen-air flame. (Courtesy of R.A. Strehlow, University of Illinois, Urbana, Illinois. Originally in R.A. Strehlow (1969).)

Fig. 4. Cellular flame in a state of chaotic self-motion. Numerical solution of Eq. (8) in $5\lambda_c \times 5\lambda_c$ square with periodic boundary conditions.

Fig. 5. One of the level curves of a steady, nearly regular flame near the stability threshold. Numerical solution of Eq. (9) in $10\lambda_c \times 10\lambda_c$ square with periodic boundary conditions; $G=2/33$.

Fig. 6. Thermal-expansion induced steady folds. Numerical solution of Eq. (18) in $5\lambda_c \times 5\lambda_c$ square with periodic boundary conditions.

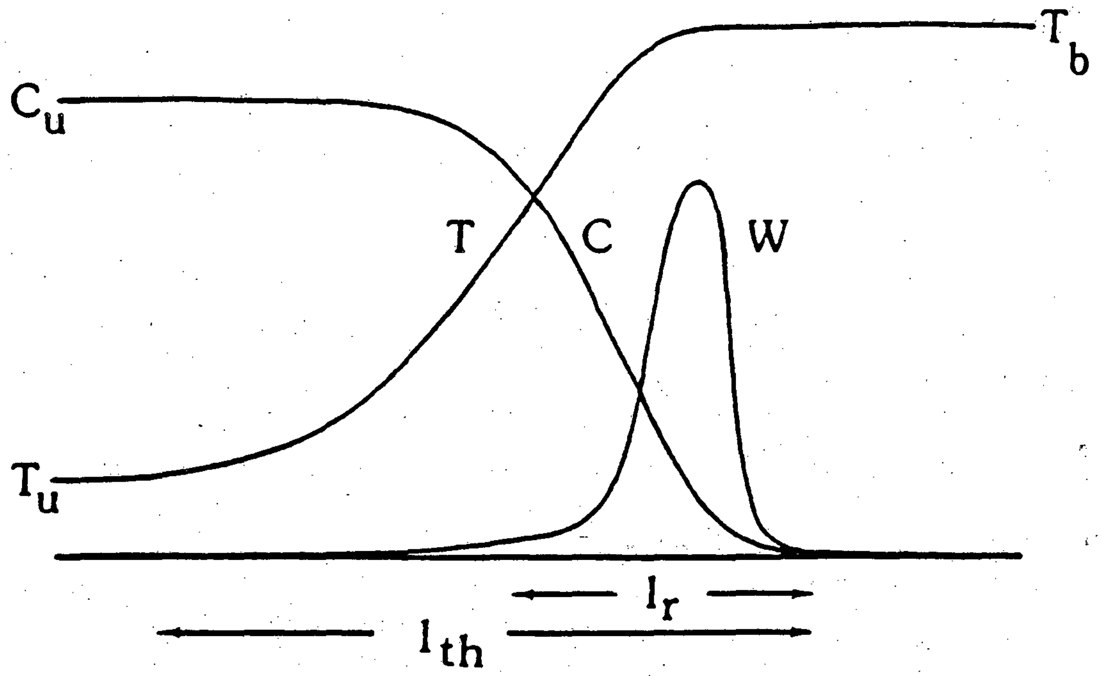


Figure 1

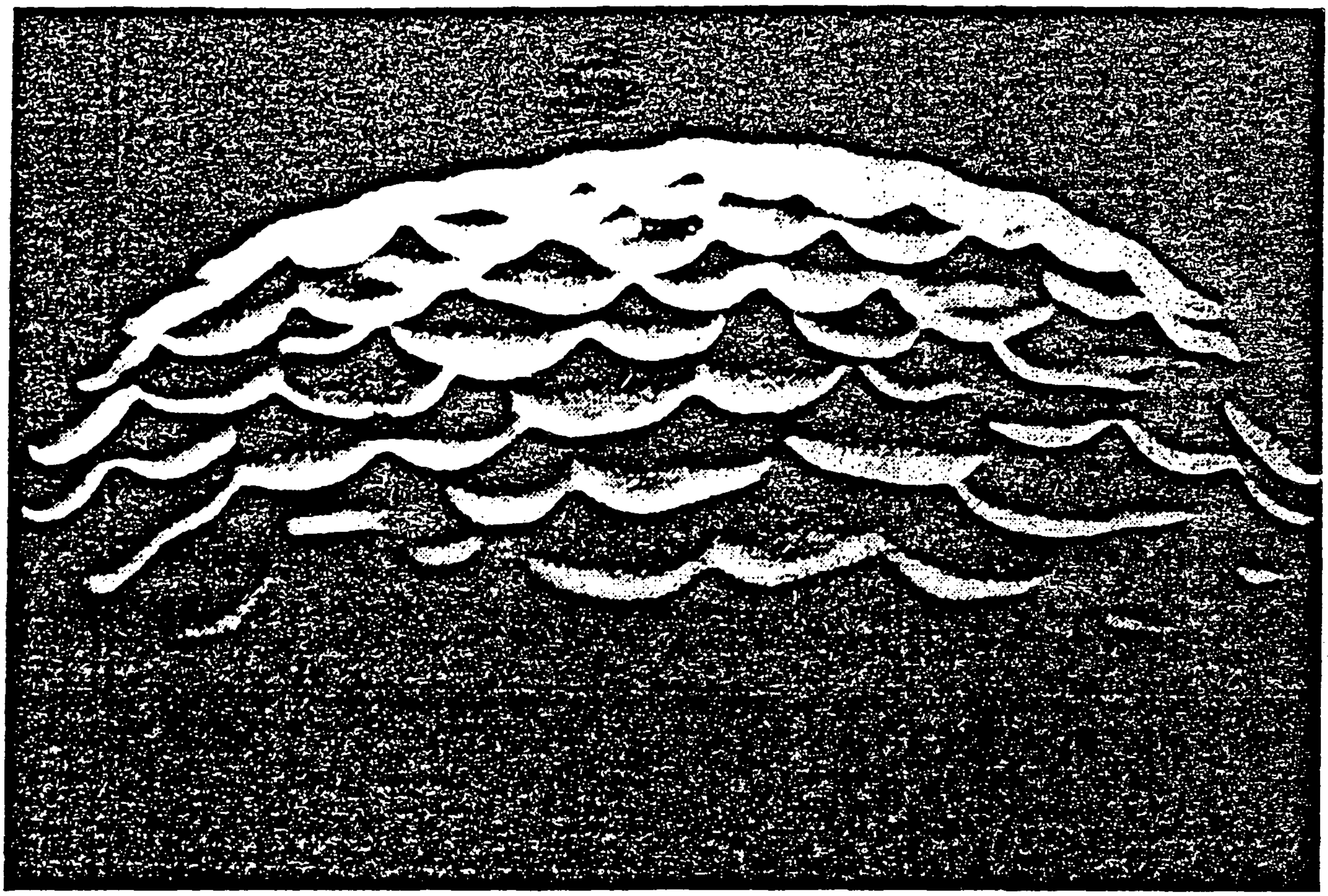
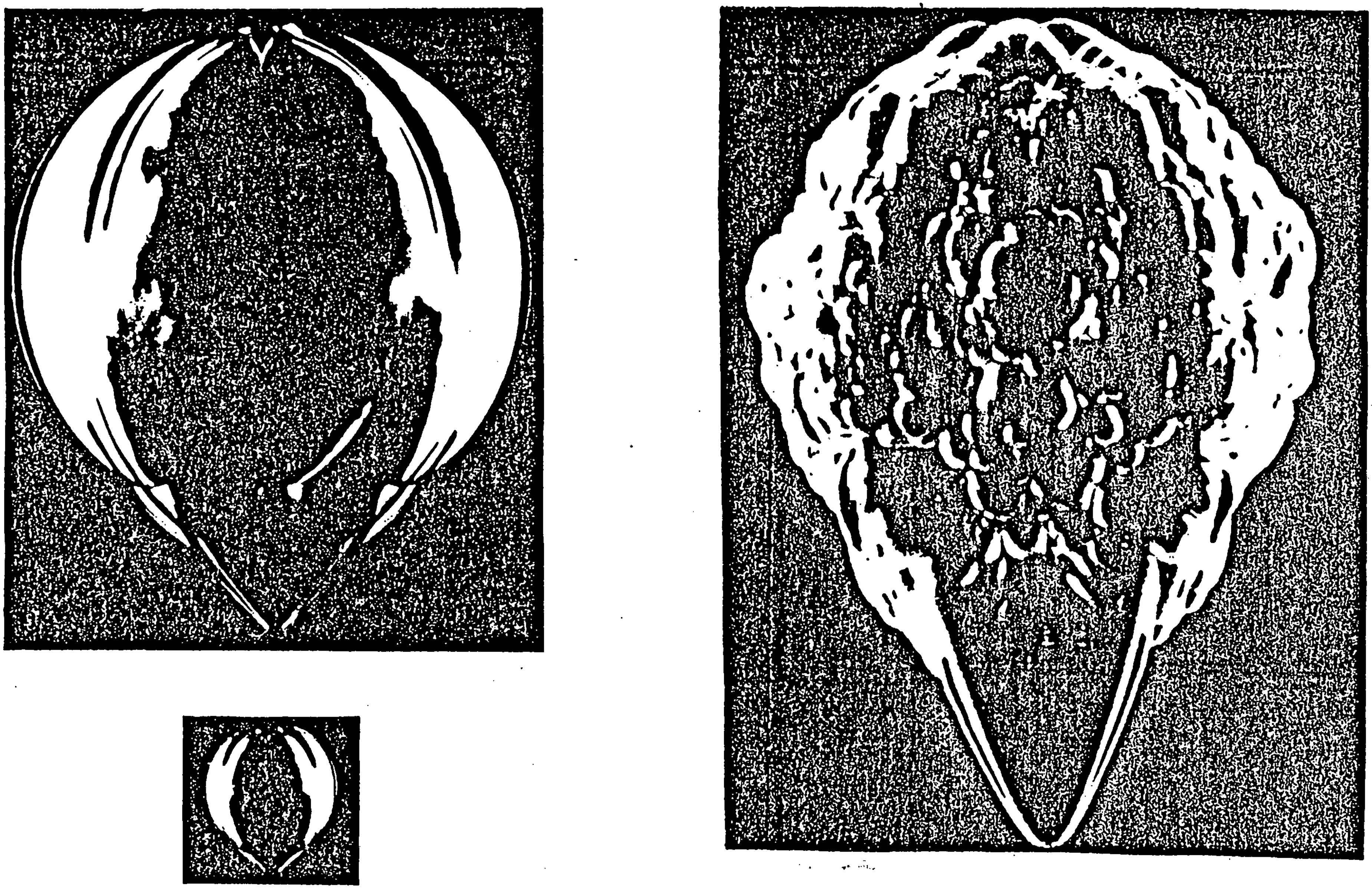


Figure 2



(a) (b)

Figure 3

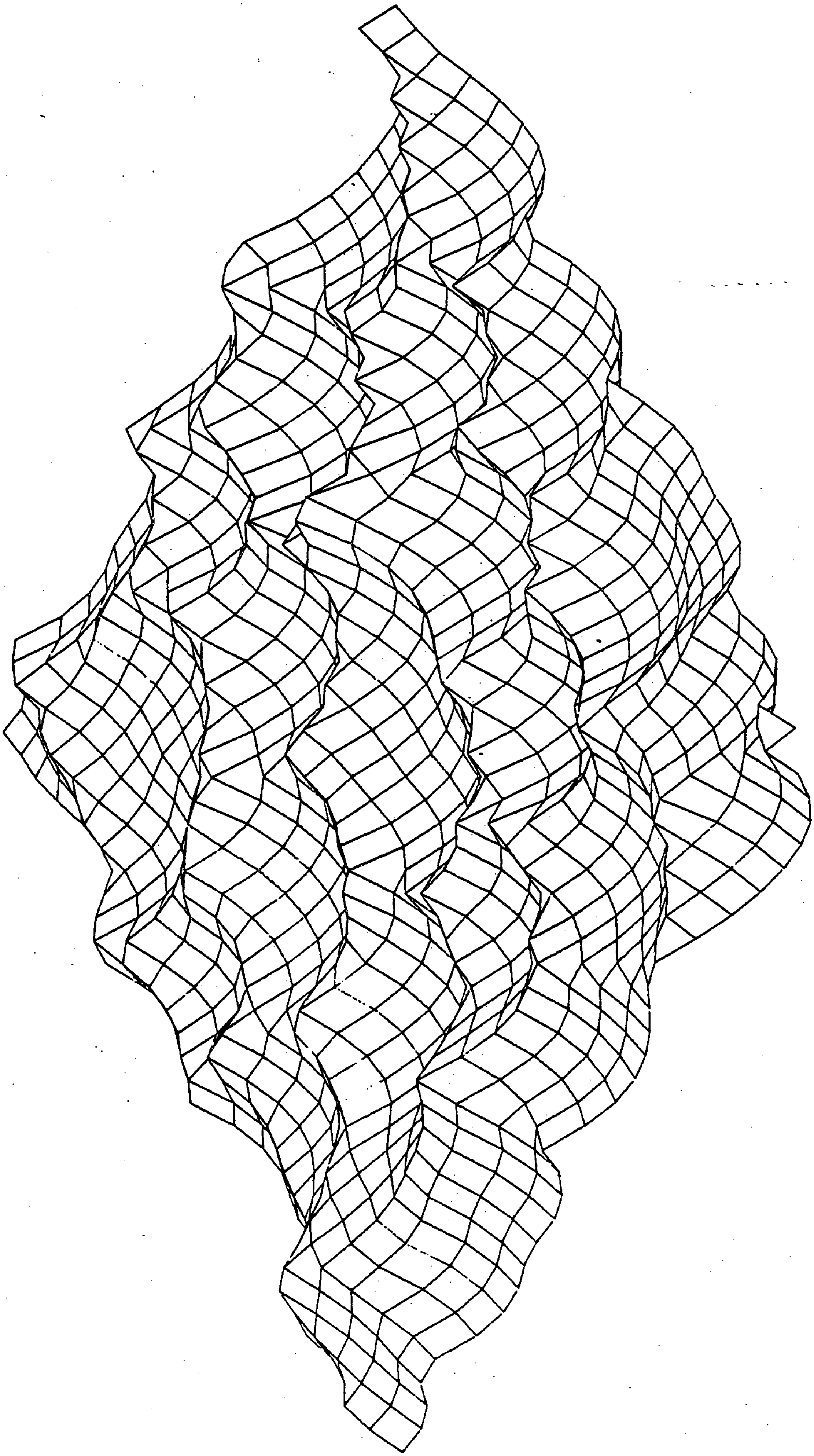


Figure 4

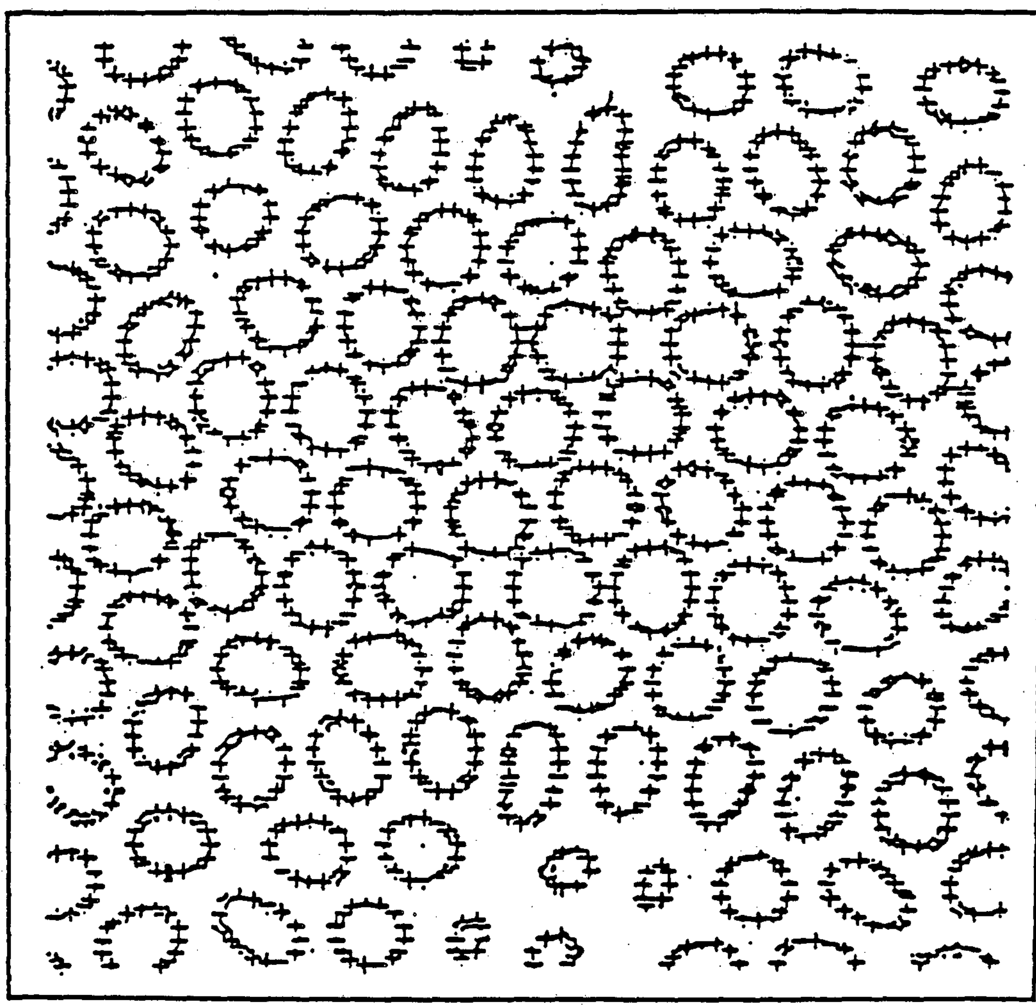


Figure 5

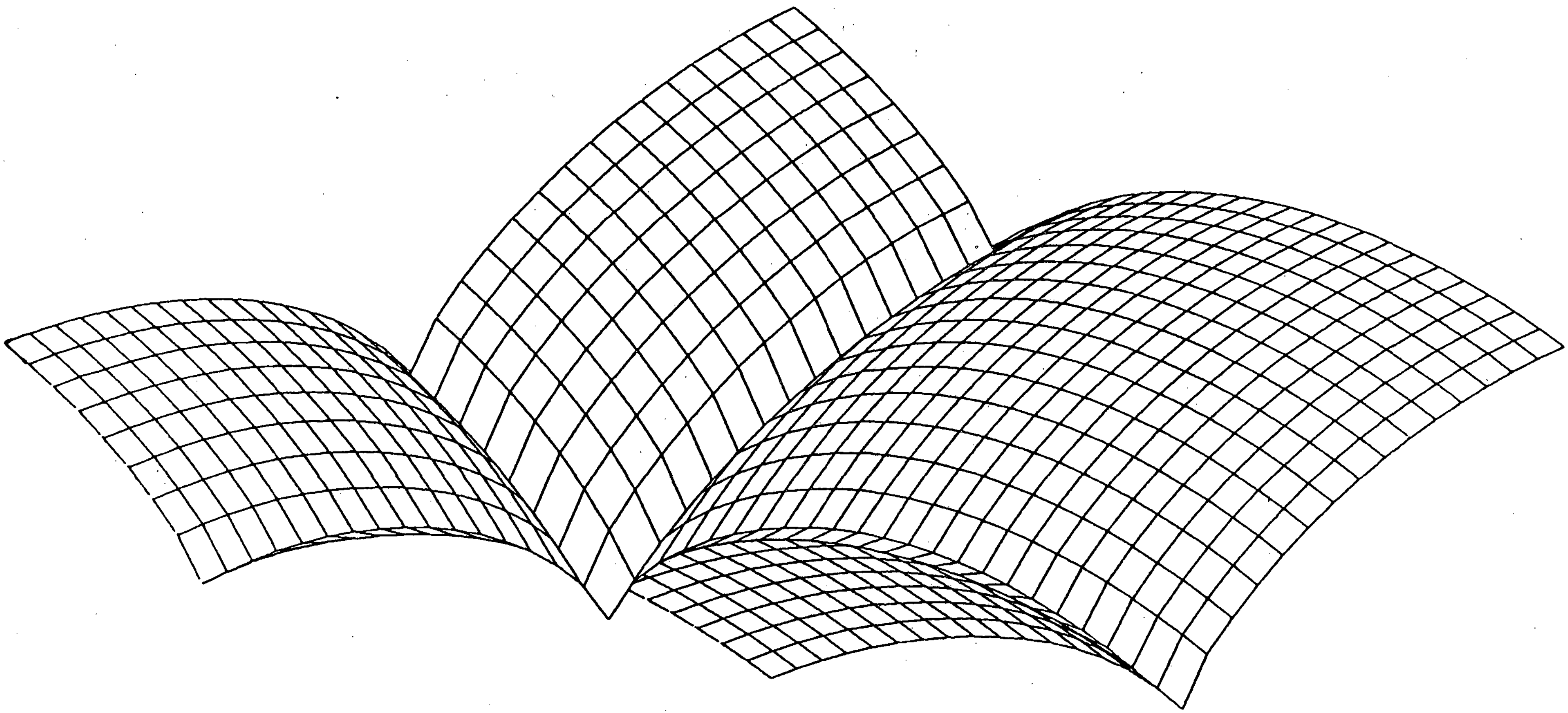


Figure 6

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