Int. J. Heat Mass Transfer. Vol. 28, No. 2, pp. 479–483, 1985
Printed in Great Britain

0017-9310/85 $3.00 + 0.00
Pergamon Press Ltd.

Dissipative structure in detonation waves

FRITZ SCHULTZ-GRUNOW
Applied Mechanics Department, Technical University of Aachen, 5100 Aachen, Templergraben 55, F.R.G.

(*Received* 12 *December* 1983 *and in revised form* 9 *February* 1984)

To Hans Liepmann's 70th Birthday

Abstract—The regular patterns which occur in detonation waves are understood with respect to their propagation but not to their initial origin. By solving the heat conduction equation for a reactive gas, one sees that order evolves from the disordered temperature distribution which is produced by any ignition. This dissipative structure is the result of the interaction of a rate and transfer process of heat. With observed reaction rates, it is found that it occurs at the end of the combustion zone in a starting detonation. The resulting heat accumulation produces the known local explosions. The distance between them is seen to be a physical quantity.

INTRODUCTION

STEADY detonation waves, which travel in combustible gases can be seen to have a regular structure which presents itself in streak photographs as an undulated wave front with specific amplitude and wavelength, with a regular pattern of striae in the burnt gas in addition to a regular pattern of crossing lines on a sooted wall over which a detonation wave had swept. The lines have a quite specific spacing and a track angle of precisely 30° [1]. It is mainly the patterns in rectangular ducts that have been observed. There they consist essentially of straight, crossing lines [2]. The same track angle and the same spacing occur in cylindrical detonations, where in consequence, the soot patterns on the plane walls of a flat combustion drum consist of logarithmic spirals. As soon as their spacing gets too large, new spirals appear in order to maintain the spacing. With spherical detonations such patterns also have been observed [1].

Current work [2] focuses on the proof of stability of a plane detonation front. A simplified model must be introduced, the 'square wave', with a finite induction zone and an infinitely thin combustion layer. It was mentioned that a finite combustion layer should be considerably more stable. Nevertheless the expected break-up of the detonation front was not confirmed. Another quite different approach was the study of transverse wave propagation. The applied acoustic ray theory must assume a sound source with ultrahigh frequency. One of the sponsors of that treatment [3] conceded that the spacing of the pattern cannot be controlled in this way 'but instead by some, as yet unresolved, finite amplitude mechanism'. A final verdict may be seen in the remark [2] that with any of these investigations 'we do not know what sort of initial perturbation may produce them'. The perturbations in the detonation front are referred to.

DISSIPATIVE STRUCTURE

The remarkable steadiness of the patterns points to a dissipative structure of the kind proposed by Glansdorff and Prigogine [4]. However at first, one has to analyse the quantities involved. The meaning of the spacing l of the pattern is disclosed by dimensional analysis which also yields less relevant dependences

$$l \sim \sqrt{a\tau}$$

where a is the thermal diffusivity and τ, the time needed to burn a fixed quantity. a is inversely proportional to pressure p, as is τ, with the usual second order reaction so that

$$l \sim \frac{1}{p}; \tag{1}$$

the dependence, in fact, has confirmation from previous measurements [1, 2]. The pressure induced is actually the partial pressure of the reactant and therefore, one should plot the measurements against this pressure, and not the total pressure, as is usually the case; thus the concentration does not appear as a parameter. For this reason, l is a physical quantity and not an eigenvalue.

A steady detonation wave consists of a strong pressure front attached to which is an induction zone, where ionization and dissociation take place followed by an exothermic recombination zone. When combustion is practically completed the so called Chapman–Jouget (C.–J.) state exists which is determined by the conservation laws of mass, momentum and energy and from reaction kinetics [5, 6]. The off-flow of the burnt gas is found to be sonic.

Special interest must be paid to the time dependence of combustion. The corresponding calculations will not be used on account of the known uncertainties about the detailed process of reaction, and it is more reliable to refer to previous experiments. Kistiakowski and Kidd [7] investigated H_2–O_2 mixtures with added inert gases in the case of a passing detonation wave. They were able to derive the overall reaction, which was shown to be of second order and in addition, an excess of O_2 appeared to act like an inert gas. This allows the assumption of stoichiometric mixtures.

NOMENCLATURE

a	thermal diffusivity [$m^2\ s^{-1}$]
c	sound speed
C_p, C_v	specific heats
D_d	detonation speed
h	delay [mm]
k	reaction rate constant
l	spacing
p	pressure
t	time [s]
T	mean absolute temperature
u	gas velocity [$m\ s^{-1}$]
U	burning rate [s^{-1}]
x, y	coordinates [mm].

Greek symbols

Δ	Laplace operator
ϑ	temperature perturbation
γ	specific heat ratio
λ	thermal conductivity
μ	burnt fraction
ρ	density.

If μ is the burnt fraction, the overall burning rate in $1\ s^{-1}$ is

$$U = -\frac{d(1-\mu)}{dt}. \tag{2}$$

With the reaction rate constant k in $m^3\ kmol^{-1}\ s^{-1}$, second order reaction is

$$U = 2k(1-\mu)^2[O_2]_0 \tag{3}$$

where subscript '0' refers to initial quantities and the square bracket to concentrations in $kmol\ m^{-3}$.

To avoid long calculations which would exceed the experimental accuracy, the mole change is neglected. This introduces a 5% deviation if 70% Argon is admixed. The solution of equations (2) and (3) is then

$$\frac{1}{(1-\mu)} = 2[O_2]_0 k\tau \tag{4}$$

which allows determination of k from the measured burning times τ.

While production of perfectly homogenous gas mixtures is no problem, any method of ignition introduces temperature disturbances. This would suggest the superposition of small perturbations ϑ to the mean temperature. As will be seen, they have a marked influence at the end of the combustion zone, where the rate of reaction is practically constant on account of its asymptotic decrease [7]. One therefore is inclined to apply the equation of heat conduction neglecting compressibility and introducing C_v instead of C_p owing to the rapid reactions to be considered. A term $U\vartheta$ has to be added which expresses the heat production by reaction. Thus

$$\frac{\partial\vartheta}{\partial t} = a\Delta\vartheta + U\vartheta.$$

Since the combustion is bound to a surface the operator Δ refers to the coordinates x, y with x parallel to the wall. The x, y-plane is parallel to the detonation front. The solution is

$$\vartheta = \vartheta_0 \sum_\alpha \sum_\beta \sin(\alpha x \pm \beta y) \exp - [(\alpha^2+\beta^2)a - U]t. \tag{5}$$

For reasons of symmetry, three adjacent intersection points of the lines of amplitudes of $\sin(\alpha x \pm \beta y)$ must form an equally sided triangle. Then $\alpha/\beta = tg\ 60°$ and $\alpha^2+\beta^2 = 1.33\alpha^2$ with

$$\alpha = \frac{2\pi}{l'} \tag{6}$$

where l' is the wavelength in the x-direction. A steady solution exists if

$$1.33\alpha^2 a = U. \tag{7}$$

This condition relates l' to the heat release in the same way as equation (1) relates the spacing l to the combustion time τ. This is as expected.

As long as the combustion is strong, and therefore U large, the corresponding steady wave-length is too small to be of relevance. At the end of the combustion zone, however, U becomes small enough so that l' may become equal to l. The steady perturbations ϑ enhance the reaction, especially at the intersection points of their amplitudes. A 1% perturbation amplitude raises the Arrhenius factor by 10% with an activation energy of 239 $kJ\ mol^{-1}$ [7] and a C.-J. temperature of 3300 K. This is equivalent to a 5% richer mixture which, in fact, is able to initiate superposed local explosions.

If one determines, with the measured soot spacing l [2], the time factor U by using equations (6) and (7) and with the burning time τ measured for $\mu = 0.75$ the constant k by using equation (4), then μ is determined by equation (3). The intermediate results of this calculation are represented in Table 1 and the obtained values of μ are listed in Table 2. One sees that the order of magnitude of $(1-\mu)$ at which the local explosions occur is a few percent. This agrees with our assumption that the perturbations ϑ only gain influence at the end of the combustion zone. There they transform the homogenous burning to uniformly spaced local explosions, which can be seen well in Soloukhin's open shutter photograph [8].

With cylindrical and spherical detonations, the specific spacing is maintained since the perturbations are present as long as heat is released, so that additional explosions originate in the same way as the first ones.

Table 1. C.-J. states ($)_2$, reaction times and reaction rate constants k

Mixture	$2H_2+O_2+0.775$ A	$2H_2+O_2+0.7$ A	$2H_2+O_2+0.5$ A
p_1 (N cm^{-2})	10.13	10.13	10.13
T_1 (K)	298.2	298.2	298.2
ρ_1 (kg m^{-3})	1.38	1.29	1.06
p_2 (N cm^{-2})	155.15	155.15	155.15
T_2 (K)	2833	3056	3355
ρ_2 (kg m^{-3})	2.22	2.13	1.72
D_d (m s^{-1})	1540	1680	1920
C_{v2} (J mol^{-1} K^{-1})	20.56	23.24	26.13
γ_2	1.404	1.385	1.32
c_2 (m s^{-1})	959	1012	1169
λ_2 (10^4 kJ m^{-1} s^{-1} K^{-1})	0.47	0.43	0.65
$[O_2]$ (kmol m^{-3}) at C.J.	0.00494	0.0067	0.011
τ (μs)	0.7	0.53	0.32
k (10^{-6} m^3 kmol^{-1} s^{-1})	578	565	574

* Without ionization and dissociation.

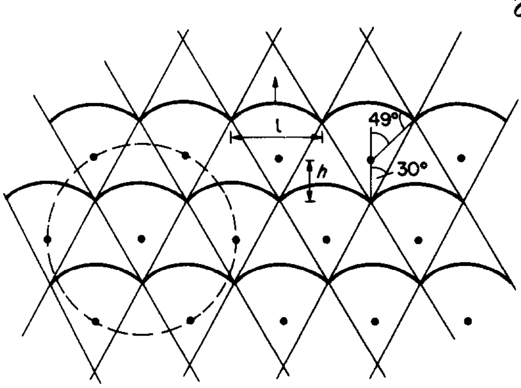

FIG. 1. Soot pattern and explosion centres.

One sees that order evolves from random disturbances by the aid of combustion which furnishes the driving energy. This is specific for dissipative structures [4].

SELF SUSTAINING GASDYNAMIC PROPAGATION PROCESS

As soon as the spacing is established and the first explosions have occurred, a self sustaining gasdynamic propagation process starts in a way such that these local explosions produce pressure waves which collide. When a critical intersection angle is established a bridge wave develops, a so-called Mach stem [1, 9] which is a typically nonlinear effect. At its ends the original pressure wave, the reflected wave and the Mach stem join so that these ends are triple points [2]. A strong shear layer develops here which, when situated at the wall, turns off soot particles thus drawing a sharp line in the soot layer of the wall. The Mach stem produces pressure pikes which ignite new local explosions. In this way a second row of explosions is produced with the same spacing as that of the dissipative structure. This process is repeated continuously. Seen from the observer on the detonation front, the lined up secondary explosions seem to jump forward and (over a distance $l/2$) to the side. Obviously the sidewise jumping must have given the impression of 'transverse waves' [9]. This process is shown in Fig. 1. The plane which is represented is the sooted wall. The solid points had been explosion centres. They are carried along with the flow with the expanding circular pressure waves until, by their collisions, new Mach stems are produced ahead of the former ones. The straight lines represent the soot pattern. Figure 1 shows that for reasons of symmetry the track angle must be 30°. Otherwise the present and past explosion centres would not have the same distance. Now, the angle of intersection at which the Mach stem originates is larger, $2\times 49°$ [10]. Therefore, in order to maintain the track angle a time delay (h) must exist for the occurrence of the explosion, which is plausible, since without delay, there would not exist a pressurized portion of gas to explode. It is interesting that this delay must occur for geometric reasons; however, h also corresponds to kinetic conditions. With l = 3 mm (Table 2), h is 0.87 mm. The

Table 2. Critical concentration $1-\mu$ for dissipative structure

Mixture	$2H_2O_2+0.775$ A	$2H_2+O_2+0.7$ A	$2H_2+O_2+0.5$ A
1 (mm)	3.0	1.7	0.9
$1.33\alpha^2\lambda$ (10^4 kJ m^{-3} s^{-1} K^{-1})	0.115	0.323	1.765
$1-\mu$	0.03	0.037	0.05

Mach stem produces the flow velocity $u = 620$ m s^{-1} [10]. The absolute velocity behind the Mach stem is (Table 1) $D_a - c_2 + u = 1200$ m s^{-1} so that there is a time delay $\tau^* = 0.7$ μs which is reasonable [10]. Since the Mach stem propagates more rapidly than the speed of sound, the local explosions are able to follow the detonation wave.

CONCLUSIONS

These two-dimensional considerations in a plane parallel to the front are restricted to ducts with a small aspect ratio. Otherwise the pressure waves must be seen as spheres so that the Mach stems which draw the soot pattern are sections of circles in a longitudinal plane normal to the wall. Additional collisions then occur inside the flow [1]. This complicates the picture, especially the side view of a detonation in interferograms, so that turbulence was supposed [11]. A closer look, however, detects a periodicity and pressure waves of the type described here. The obvious cause of the impression of irregularity is that the local explosions are seen one above the other. Unfortunately, a square duct was used. Streak photographs in narrow ducts do not reveal turbulence at all but a regular wavy pattern which could even be derived with the concept of local explosions [1].

Such patterns only appear in steady detonations. Indeed, overdriven detonations are unstable since subsonic off-flow allows disturbances to reach the front and feeble detonations are not steady since the distance between shock and combustion layer varies and the pressures in the latter are low. White's 'laminar' detonation [12] is presumably such a nonsteady detonation. This is indicated by the experimental set up and by White's context.

REFERENCES

1. F. Schultz-Grunow, Microstructure of gaseous detonation waves, Rep. 6/71, Ernst Mach Institute, Freiburg i. Br., Eckerstrasse 4, W. Germany (1971).
2. W. Fickett and W. C. Davis, *Detonation*. University of California Press, Berkeley (1979).
3. R. A. Strehlow, R. Liangminas, R. H. Watson and J. R. Eyman, Transverse wave structure in detonations, Eleventh Symposium (International) on Combustion, pp. 683–692. The Combustion Institute, Pittsburgh (1967).
4. P. Glansdorff and I. Prigogine, *Structure, Dissipation and Fluctuations*, John Wiley, New York (1971).
5. C. L. Eisen and R. A. Gross, *Theoretical Calculations in Gaseous Detonations, Combustion and Flame*, Vol. 4, pp. 137–147. The Combustion Institute, Pittsburgh, Pennsylvania (1960).
6. J. H. Schaefer, Detonation and deflagration in explosive gas mixtures, rep. 3/73, Ernst Mach Institute, Freiburg i. Br., Eckerstrasse 4, W. Germany (1973).
7. G. B. Kistiakowsky and P. H. Kidd, A study of the reaction zone by gas density measurements, *J. chem. Phys.* **25**, 824–835 (1956).
8. R. I. Soloukhin, *Shock Waves and Detonations in Gases*. Mono Book, Baltimore (1966).
9. R. A. Strehlow and R. E. Maurer, *Transverse Waves in Detonations I*, AIAA-J, Vol. 7, pp. 323–328. American Institute for Astronautics and Aeronautics, New York (1969).
10. F. Schultz-Grunow, Generation of the patterns in gaseous detonations, *Proceedings Thirteenth International Symposium on Shock Tubes Shock Waves*, pp. 352–357. State University of New York Press (1982).
11. J. A. Fay, The structure of gaseous detonation waves, *Eighth Symposium on Combustion*, pp. 30–40. Williams and Wilkins, Baltimore (1962).
12. D. R. White, Detonation wave structure near the hot boundary, *Physics Fluids* **6**, 1011–1015 (1963).

STRUCTURE DISSIPATIVE DANS LES ONDES DE DETONATION

Résumé—Les configurations régulières qui apparaissent dans les ondes de détonation sont comprises tant que l'on considère leur propagation mais pas leur tout début. En résolvant l'équation de conduction thermique pour un gaz réactif, on voit que l'ordre évolue à partir d'une distribution décordonnée de température qui est produite par une ignition. Cette structure dissipative est le résultat de l'interaction d'une vitesse et d'un transfert de chaleur. Avec les vitesses de réaction observées, on trouve que cela apparait à la fin de la zone de combustion dans une détonation commençante. L'accumulation de chaleur résultante produit les explosions locales connues. La distance entre elles est une grandeur physique.

DISSIPATIONS-STRUKTUR IN DETONATIONSWELLEN

Zusammenfassung—Die regelmäßigen Muster, die in Detonationswellen auftreten, werden bisher nur im Hinblick auf die Ausbreitung verstanden, nicht jedoch im Hinblick auf ihre Entstehung. Durch die Lösung der Wärmeleitgleichung für ein reagierendes Gas sieht man, daß sich aus der ungeordneten Temperaturverteilung infolge einer Entzündung eine Ordnung entwickelt. Diese dissipative Struktur ergibt sich aus dem Zusammenwirken von Wärmefreisetzung und Wärmeübertragung. Mit Hilfe der beobachteten Reaktionsgeschwindigkeiten wurde herausgefunden, daß der Beginn der Detonation am Ende der Reaktionszone auftritt. Die Ansammlung von Wärme, die sich dabei ergibt, führt zu den bekannten örtlichen Explosionen. Ihr Abstand erweist sich als eine physikalische Größe.

ДИССИПАТИВНАЯ СТРУКТУРА ДЕТОНАЦИОННЫХ ВОЛН

Аннотация—Развивающиеся в детонационных волнах регулярные структуры легко поддаются описанию при их распространении, но не в самом начале зарождения. Из решения уравнения теплопроводности для реагирующего газа можно видеть, что упорядоченная структура развивается из неупорядоченного распределения температуры, возникающего при воспламенении. Диссипативная структура такого типа является результатом взаимодействия скорости реакции и процесса теплопереноса. Найдено, что при исследуемых скоростях реакции такая структура образуется в конце зоны горения при зарождении детонации. В результате аккумуляции тепла происходят локальные взрывы, расстояние между которыми представляет собой физическую величину.