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# Recent work on gaseous detonations

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**Abstract.** The paper reviews recent progress in the field of gaseous detonations, with sections on shock diffraction and reflection, the transition to detonation, hybrid, spherically-imploding, and galloping and stuttering fronts, their structure, their transmission and quenching by additives, the critical energy for initiation and detonation of more unusual fuels. The final section points out areas where our understanding is still far from being complete and contains some suggestions of ways in which progress might be made.

**Key words:** Review, Detonation, Unsteady waves, Gaseous explosion, Damage

## 1 Introduction

Now seems to be an appropriate time to review progress on my suggestions (Nettleton 1987) for further work in the field of gaseous detonations. I make no apology for my focus on safety aspects. Recall that Dixon (1893, 1903) started work in this area, following a gaseous detonation in a town gas main in London. My introduction was through a series of explosions during start-up and shut-down of turbulent flows of suspensions of coal dust in air in pipelines, running between mills on the basement floor and burner nozzles in the walls of modern (typically 500 MW) pulverized-fuel fired plant. During normal operation, concentrations of dust in these are above the upper flammability limit, so that unsafe conditions are of very short duration. A wide variety of plant may be threatened by the possibility of a detonation either in the gaseous phase, by a self-decomposing material, by a mixture of fuel and oxidant or by a flammable dust suspension. It is often possible to design to contain the effects of an unreflected detonation; for instance, straight runs of pipelines can, without excessive cost penalties, be made thick enough to cope with the results of an unreflected detonation. Problems, however, will occur at any bends. The German hydrocarbon-synthesis industry in the 1940s adopted a different approach, seeking to prevent the detonation of acetylene, either pure or diluted by inert additives, by tightly packing all containment and designing to cope with any resulting deflagration. This was a remarkable feat, with packed pipes and vessels withstanding the results of bombing attacks.

Dust suspensions in power stations flow through cast-iron pipelines, designed to be thick enough to withstand erosive wear of particles of pure silica, and thus, in their unworn state, are capable of containing an unreflected detonation. However, bends are often made of more ductile mild steel to prevent fragmentation under the influence of the higher pressure as the front reflects. It is common practice with explosive dusts to build plant and its containment as lightweight as possible, and rely on a combination of fitted vents and failures of the structure to prevent the formation of a detonation. Clearly, information on the behavior of light and massive components of both ductile and brittle materials are required both to design plants and to predict from any resultant damage, whether they have been subjected to a detonation.

In the past decade there have been a number of reviews of detonations, but only two of these have concentrated on safety aspects. For instance that by Clarke (1989) is strongly biased towards a mathematical analysis of the acceleration of flames fuelled by a single step reaction. There must be questions about the inaccuracies involved in treating the chain reactions of both gaseous oxidation and decomposition of self-combusting fuels in terms of a single rate constant, probably more so for the former than the latter, where there is a significant shortening in the number of steps. In this context, explosions of dust-oxidant systems are generally believed to be homogeneous. Accepting the uncertainty in chemical kinetics, the paper forms an ideal introduction to analytical techniques for predicting the transition from deflagration to detonation (DDT).

Lee (1991) gives a complete rehearsal of the reasons compelling a belief in the multi-dimensional nature of detonation fronts. I have looked at the nature of detonation fronts in ram accelerators, where detonations are driven between a projectile and its containment walls in mixtures of unusual composition at pressures of up to 25 bar in atmospheres of unusual composition (Nettleton 2000). The paper gives details of the interaction of shocks with walls and reaction zones together with details of the interaction of a planar detonation with a surrounding medium, which can either be inert or of different composition.

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Of the two papers dealing more directly with safety concerns, Gelfand et al. (1991) show that much valuable information is still derivable from uni-dimensional treatments. In particular, Russian work following on from that of Zel'dovich for uni-dimensional waves with heat losses to walls or to inert or reacting particles in the flow is comprehensively covered. Prior to this many of the reports had only been available in Russian. The most recent by Moen (1993) covers DDT in clouds of fuel in air. The costs and problems in siting and choosing suitable instrumentation for studies of explosions of large, say greater than 50 m, flattened and non-hemispherical clouds with fuels much denser than the surrounding air are considerable, so that much depends on the scaling laws chosen to extrapolate from small-scale tests.

The present review assumes familiarity with the contents of these earlier ones and goes on to discuss safety aspects of confined and unconfined fronts. In particular, sections deal with progress in understanding processes such as shock diffraction and reflection, DDTs, hybrid detonations, spherically imploding fronts, galloping and stuttering waves, front structure, transmission of the wave, the use of additives to quench detonations, those in less common fuels and critical energies for their initiation.

## 2 Influence of containment on detonation structure

The explosion in the nuclear plant at Three Mile Island has resulted in a wide range of studies of the reactivities of mixtures of oxygen, nitrogen, steam and hydrogen, see for instance Tieszen (1993). Particularly notable is that by Boyack et al. (1993) who start with the proviso that the components are likely to be sufficiently massive for their associated time constants to be very much longer than the rise-time and probably the duration of the pressure pulse. It should be noted that is by no means generally true for other types of plant. They report on some of the mechanisms leading to enhancement in pressure, such as DDT or the presence of thin layers of inert materials. Whilst detonation velocities can be reduced by their presence (Gelfand et al. 1991) the momentum, depending on mass, may not. Their analysis shows that edges and corners can enhance pressures predicted from normal reflection and that peak pressures are a function of equivalence ratio and the thickness of the layer of inert gas.

Extending earlier theoretical studies in the application of the Chester-Chisnell-Whitham (CCW) relationship for detonations in convergent channels, Bartlmä (1990) and Li and Ben-Dor (1998) examined detonative flows over wedges, correcting the strong shock approximation to allow for a change in ratio of specific heats across the front. The implicit assumption here appears to be that the reaction zones assume a similar shape to that of the leading front. A comparison of theory with experiment suggests that the optimal agreement with experiment is obtained by assuming a degree of overdrive of 10%. Furthermore, there remain questions about the effects of cell size on the trajectory of the triple point. Edwards et al. (1984) show these are of particular importance at low angles of the wedge. A somewhat similar paper by Li et al. (1997) compares theoretical and experimental results for the angle of the oblique reflection of detonation waves in diluted and undiluted mixtures of oxyhydrogen, assuming coupling of lead front and reaction zones. Again the theoretical results are very sensitive to the assumed degree of overdrive.

The critical diameter problem for the transition from planar to spherical detonation continues to excite attention and has recently been reviewed by Lee (1996). Jones et al. (1995) have produced a mathematical model to account for the effects of cell spacing in oxyhydrogen mixtures diluted by argon on transmission in two-dimensional expansions. Again, Pantow et al. (1996) have examined experimental and theoretical results for the propagation of detonations in oxyhydrogen mixtures through sudden expansions. They obtained close agreement with experiment using the random generation of numerical noise to produce the transverse fronts. They do not comment upon the mechanism whereby certain fronts grow whilst others decay. Gamezo et al. (1998) in a study of planar shocks also used numerical noise to impose disturbances and show that the front is unstable to perturbations with wavelengths greater than one to two reaction zone half-widths. The noise was found to govern the initial process, but not the final spacing of transverse fronts. An increase in the energy of activation leads to the formation of a more irregular structure. Jones et al. (1996) have taken a somewhat different approach in their theoretical modeling of an expanding front, complimenting that of Pantow et al. (1996), but lack an experimental validation of their analysis.

## 3 The transition from deflagration to detonation

This is an area which has received much attention over the last decade. It is one that also creates some problems with a number of papers impinging on topics with which I have chosen to deal separately. Possibly an ideal starting point is the paper by Chue et al. (1993) giving a numerical analysis of the structure of fronts travelling approximately at the speed of sound in the hot combustion products. Interesting papers by Zhang et al. (1992, 1998) return to the Craven and Greig (1968) postulate of maximum pressures being generated by a transition process which produces a precursor front with a reflected shock travelling back through this zone. Their theoretical predictions are compared with measurements of pressure in an end slug of aluminium dust in a mixture of acetylene in air. Here peak pressures were up to twice those for the homogeneous explosive mixture. It cannot be too strongly emphasised how important inhomogeneities in the mixture are in determining final pressures.

Ciccarelli et al. (1994) have extended previous work on DDT in hydrogen-steam-air mixtures at ambient temperatures up to 650 K, about that anticipated to have

occurred in the Three Mile Island explosion, using tubes containing a number of orifice plates. These proved to be successful for mixtures of composition such that cell spacing was close to the diameter of the orifice.

Saurel et al. (1992) have shown that distances for DDT are reduced when the initiation source is a hot plasma. They give a numerical treatment for laser initiation in a two phase medium, emphasising the care which must be taken in the choice of an appropriate equation of state; see also Fujiwara et al. (1994) for further details of computational analysis of laser supported detonations in homogeneous mixtures.

Johnson et al. (1996) have examined the influence of two flame inhibitors, both halogenated hydrocarbons, on transition distances in mixtures of methane, propane and acetylene with air. In each case the velocity of the flames in obstacle-filled tubes was reduced and, with a sufficient amount of inhibitor, quenched.

A series of papers from Moscow State University on theoretical and experimental studies of DDT for homogeneous and droplet dispersions in air have recently appeared, see Smirnov and Tyurnikov (1994, 1995), Smirnov et al. (1995, 1997), Smirnov and Panfilov (1995). They extend the range of fuels tested to, for example, $n$-methylenecyclopropane and $n$-decane (see section on less usual fuels). Smirnov et al. (1997) give details of rates of wall loads to be expected in the event of the reflection of the detonation formed under different scenarios.

Following Teodorczyk's (1995) work on the role of instabilities in promoting turbulence and thus flame acceleration resulting in DDT, Khokhlov et al. (1997) postulate that transition occurs via the formation of gradients produced by a combination of turbulent mixing and local quenching of the flame. A further paper by Khokhlov et al. (1999) gives a numerical approach to the role of shock-reaction zone interactions in turbulent flows. Obara et al. (1996) present the results of a high speed schlieren study of DDT in oxyhydrogen mixtures with different degrees of dilution by nitrogen in obstacle-filled channels. Local overdriven fronts travelling at up to twice the C-J velocity were observed. Chue et al. (1995) have conducted a theoretical and experimental investigation of the transition in oxyacetylene mixtures diluted by argon. In order to obtain fast deflagrations, wire screens were used to suppress any transverse structure. However, transverse pressure waves rapidly reformed, eventually to produce transverse reaction fronts, following their interaction with the leading shock. Teodorczyk et al. (1988) have also photographed the propagation of quasi-detonations in mixtures of hydrogen, ethylene and propane with oxygen in obstacle-filled rectangular tubes.

Carnasciali et al. (1991) have studied the initiation of detonation in ethylene, acetylene, propane and hydrogen in mixtures of oxygen and nitrogen by a turbulent jet of hot gases (see also Achasov et al. (1995) in Sect. 10). They concluded that there is a critical ratio of size of jet orifice to cell width that is comparable with that of 13, the ratio of tube diameter to cell width, necessary for the initiation of spherical fronts. It is unfortunate that Khokhlov et al. (1999) do not appear to make any use of the data from these experimental studies.

Brown and Thomas (1999) have measured ignition delays in the temperature range 1073 to 2211 K in both ethylene and propane diluted by varying amounts of either argon or nitrogen and combined these with spark schlieren photographs of shock reflection, ignition and transition to detonation. They show that only a complex, and inordinately expensive to compute, kinetic scheme involving some 600 reaction steps predicts accurately delays to ignition.

Two related papers (Frolov et al. 1994; Noscov et al. 1995) deal with numerical investigations of the effects of auto-ignition in the wall boundary layer of homogeneous mixtures. The resultant flame can propagate at close to the local velocity of sound, leading to strong coupling between energy release and local pressure fronts.

Starikovskii (1996) has examined the ignition and transition process in nitrous oxide/carbon monoxide/hydrogen mixtures diluted by helium. Undiluted mixtures have some interesting characteristics, including the possibility of super-equilibrium concentrations of carbon dioxide and the potential existence behind reflected shocks of gas-dynamic perturbations leading to DDT. Both theory and experiment reveal that single and two-phase modes occur and are controlled by a combination of the total heat release and the ratio of induction and exothermic reaction times.

Recent papers on DDT in dust suspensions include that by Klemens et al. (1994) who found that the presence of inert particles of quartz had little influence on transition distances. However, they produced a linear decrease in flame velocity and finally extinction. Li et al. (1995a,b) in two very similar papers deal with the transition of dust layers deposited on walls of tubes. Their principal finding is the importance of inhomogeneities in concentration in determining DDTs.

Corn-starch is a frequent choice for the combustible dust, see for instance Zhang and Grönig (1989, 1991) who studied transition processes in both oxygen and air. They give further results in a subsequent combined experimental paper (Zhang et al. 1992). They acknowledge the importance of non-uniformities, and make every attempt to reduce these, as far as possible and succeeded in producing runs with a deviation of less than 0.5% in a series of 15 experiments in a 140 mm diameter tube with a ratio of $L/d$ of 124. A numerical analysis by Hayashi et al. (1994) of two-phase detonation processes in suspensions of starch in air suggests that a one-step reaction mechanism suffices to produce satisfactory predictions of C-J velocities and distances for ignition and transition to detonation. However, they appear only to validate their velocities with results from some unspecified source (a referee graciously informs me that they used the data of Zhang and Grönig 1989, 1991).

A further paper on dust deposits by Fan and Li (1996) deals with wall deposits of 15 $rm\mu$m particles of aluminium and two condensed explosives, RDX and HMX, and is mainly concerned with differences between theoretical and experimental C-J velocities. In a combined theo-

retical and experimental study Fedorov et al. (1996) also chose suspensions of aluminium dust, this time in oxygen. Alexander et al. (1993) describe experiments on various dusts, both in suspensions and in wall layers, using both mild and severe forms of ignition. For suspended dusts both sources result in the formation of a detonation. However, with layers of dust mild sources are more successful than are blast waves. The experiments took place in three forms of confinement, a vertical tube of moderate length, a larger diameter horizontal tube and an explosion gallery with ignition via a flame in a methane/air mixture. Dahab et al. (1989) obtained transition in coal dust-oxygen mixtures in a vertical tube when the ignition source was of low energy. Finally, Korobeinikov (1993) has produced a theoretical analysis of the important parameters governing the onset of detonations in suspensions of dusts and pointed out the problems in formulating laws governing scaling from small to large scale detonations, particularly unconfined ones.

## 4 Hybrid detonations

The hybrid term refers to a combination of fuels, generally particles with a more detonable gas. However, I am presently extending its use to cover dust cloud containing other forms of sensitisers. In a pair of papers Veyssiere and Khasainov (1995) and Khasainov and Veyssiere (1996) set out to examine the nature of fronts in suspensions of aluminium powder in oxyhydrogen mixtures. The first uses a ZND model of the front with heat losses to predict the existence of three regimes, pseudo-gas, single and double detonation fronts. The second, an experimental paper searches for these and concludes that the first decays to a single front while the latter two appear to be stable. A more recent paper by Veyssiere et al. (1999) deals again with clouds of starch in oxyhydrogen mixtures. The theoretical analysis involves the particles heating to an ignition temperature of 700 K in oxygen, close to the value found by Nettleton and Stirling (1967) for coal particles in shocked oxygen, followed by the production of a homogeneous flame. Discrepancies in velocities in the various studies of clouds of starch outlined in the previous section are attributed to changes in particle size, dimensions and layout of the tubes.

Wolinski et al. (1996) demonstrated that hybrid detonations of oats dust are achieved in methane/air mixtures for methane concentrations of 8.5 to 14.5% and dust concentrations not greater than 0.1 kg/m$^3$. Higher loadings of dust act as a suppressant. Finally, Tulis (1989) describes how unconfined detonations of aluminium powder in air can be generated in the presence of 5% nitroguanidine acting as a sensitiser. As far as I am aware, this is the first report of unconfined detonations of dusts in air.

## 5 Spherically-imploding fronts

Two papers by Terao and Wagner (1991), Terao et al. (1995) describe the construction of an apparatus producing imploding fronts with temperatures between $10^7$ and $10^8$ K at the focus. The extreme precautions in construction of the apparatus required to ensure symmetry means that such events are not likely to occur in normal plant.

## 6 Galloping and stuttering fronts

These terms imply the continuous cycling of fronts from velocities well in excess of the C-J value to one considerably below it. Experiments examining such phenomena must be carried out in straight tubes of at least tens of meters in length to ensure the existence of their cyclic nature. When numbers of sharp bends are present, for instance in polymerizing plant for the production of polyethylene, distances may be sharply reduced. However, it is difficult to visualize such conditions occurring frequently in plant.

Dupre et al. (1986, 1990, 1991) and Lee et al. (1995a, 1995b) in a series of experiments, using different monitoring techniques, demonstrate that the phenomena can occur for a wide range of hydrocarbons in stoichiometric mixtures with both oxygen and nitrous oxide. Ishii and Grönig (1998) showed that the phenomenon is not confined to hydrocarbons, but can also occur with low pressure mixtures of oxyhydrogen, so that the phenomena cannot be accounted for by the more complex reaction schemes of the hydrocarbons.

## 7 Structure of fronts

Before discussing recent papers in this field, I should draw attention to the availability of much experimental data on spacing of fronts on the internet. Valuable addresses are http://www.galcit.caltech.edu/∼mikek/detn_db and http://www.galcit.caltech.edu/detn_db/html/, see Kaneshige and Shepherd (1997).

Of late there has been much work on numerical modelling of confined detonations in both homogeneous and dusty gases. For example, Lefebvre and Oran (1995) have examined shock structures in oxyhydrogen mixtures heavily diluted by argon. They suggest that the degree of release of energy is concentrated close to the triple point and its level increases as a triple point approaches another or the wall. From the same school, Williams et al. (1996) numerically model how, in the absence of wall losses, the complexity of structure of transverse waves increases with a change from 2 to 3 dimensions, with two perpendicular modes accounting for the slapping wave frequently observed in experiments. Further, the interaction of these waves produces the vortex field which entrains unburnt mixture behind the Mach stem. Another theoretical paper by Oran et al. (1998) uses a detailed kinetic scheme to demonstrate that two-dimensional models indicate the possibility of a second release of energy two thirds along the cell length of detonations in oxyhydrogen mixtures diluted by argon.

Fujiwara and Washiku (1994) have also studied the structure of fronts in oxyhydrogen mixtures using a simplified kinetic scheme. For spinning waves, they propose

that the diameter of the tube should lie between 1.8 and 3.4 cell widths. Bauer et al. (1986) examined the influence of initial pressure using an optical technique to measure cell lengths in three fuels, propane, ethylene and methane, mixed with oxygen and nitrogen. The most reliable results were for pressures between 3 and 50 bar in propane air mixtures, from 1 to 10 bar in mixtures of ethylene, with oxygen and nitrogen and from 0.5 to 15 bar for diluted oxymethane mixtures. They also show that a constant of 29 relates ignition distances with cell lengths throughout the pressure range.

Uphoff et al. (1996) tackle numerically the difficult problem of the structure of detonations in the oxidation of carbonaceous materials in oxygen. Together with the combination of difficulties imposed by the choice of appropriate heat and mass transfer relationships, the existence of complex chemical kinetics might be anticipated. Experimental verification also poses problems, both from the viewpoints of constant shape, size distribution and uniformity in dispersion.

Stewart et al. (1996) in a somewhat contentious paper, see questioners' comments, derive explicit criteria for cell spacing and aspect ratios for weakly curved waves in media with high activation energies. Their theory is based on the formation of pulsating waves leading to cellular instabilities. It seems doubtful that chain reactions, involved in most oxidation processes, correspond with their proviso.

Beeson et al. (1991) have given experimental results for cell sizes of fronts in $n$-hexane and commercial jet fuel in a nitrogen-oxygen mixture over the range $0 < x < 3.8$, where $x$ is the ratio of nitrogen to oxygen at 300 K. Tieszen et al. (1991) give experimental and theoretical results for cell sizes in some 20 hydrocarbons with significantly different chemical characteristics. Surprisingly, for a standard composition they vary only from 40 to 50 mm, within the experimental error between two competent observers of soot tracks.

Stamps and Tieszen (1991) and Ciccarelli et al. (1994) in response to the explosion at Three Mile Island have determined cell sizes in mixtures of hydrogen, air and diluents. Both deal with the effects of both initial pressure and temperature on spacing. Terao and Motoyama (1992) extend measurements in the influence of temperature on detonation velocities in oxypropane over the range from 400 to 700 K.

Tulis et al. (1993) working with aluminium powder suspensions in air sensitised by the addition of RDX demonstrate that it is possible to obtain both single headed and multiple fronts. As with their paper on unconfined fronts (Sect. 4), this is the first paper to report upon multi-headed fronts in clouds of dust in air. Their experiments also revealed significant differences in the performance of atomized aluminium, less than 5 μm, and flaked aluminium, equivalent to spherical particles less than 1 μm in diameter.

Nettleton and Teodorczyk (1992) endeavor to estimate the characteristic size of a model scramjet which, were mixing of air and hydrogen sufficiently fast, would be capable of supporting a standing detonation. Again on oxyhydrogen mixtures Ohyagi et al. (1992) used pressure measurements, relating falls in pressure and Mach number with the development of size of cells. Lastly, Lee et al. (1995b), in an attempt to remove the uncertainties associated with individual interpretations of widths of cells, have investigated the use of digital analysis of smoked foils.

## 8 Transmission of fronts

For convenience this section is divided into transmission through concentration gradients and through roughened or obstacle-filled containment. Recent theoretical papers include that of Voronin and Mitranov (1991). An early paper paying great attention to any mixing occurring between filling and firing a shock tube was that by Thomas et al. (1991). It demonstrated that improvements can be made to the Paterson (1953) model of refraction of the shock by taking into account the Taylor expansion and losses to the walls. Kuznetsov et al. (1997) examined the fate of waves travelling from stoichiometric oxyhydrogen into an inert medium; in this case, either argon, helium, nitrogen or carbon dioxide. The effectiveness of quenching was found to be proportional to the molecular weight and inversely proportional to the specific heat ratio of the inert material. Kryuchkov et al. (1996) in a theoretical paper examine the propagation of a detonation of a width which increases linearly under the influence of a decreasing reaction rate. The critical conditions for the gradient in reactivity is deduced by solving the continuity equations for a one-step Arrhenius kinetic model. Their prediction for moderate values of the effective activation energy suggest the distance for failure is of the order of 10 cell lengths.

Teodorczyk and Lee (1995) extended by Teodorczyk and Benoan (1996) describe experiments on the role of wall layers in attenuating transverse waves and quenching fronts. A further paper from Lee's group, Makris et al. (1995) shows that results for the influence of a porous structure in attenuating fronts can be correlated by the empirical relationship $V/V^* = 1 - 0.35\log(d\#/d)$, where $d\#$ and $d$ are the critical tube diameter, representing mixture sensitivity and the pore size.

Teodorczyk and Thomas (1995) in an experimental study explain how acoustically absorbing walls and air gaps quench and re-initiate detonations. Lyamin et al. (1991) review and extend Russian work on hydrogen, acetylene and propane mixtures with air and oxygen in porous media. For the latter, only C-J fronts were found to be stable in all media, when the mixtures are at or close to stoichiometric. The different regimes experienced with mixtures that are far from being stoichiometric and at pressures well below the critical value are described.

## 9 Quenching by additives

Two papers deal with experiments on the effectiveness of halogenated hydrocarbons in quenching established detonations (Lefebvre et al. 1995; Evariste et al. 1996). Of

those tried trifluorobromomethane was the most efficacious. Thomas et al. (1990) found that water sprays were effective, providing that losses to the droplets exceeded the Schelkin criterion of 10%. Segev et al. (1988) studied detonations travelling through foams of oxyethylene in water, showing how the composition of the medium affects both bubble size and DDT distances. For fuel-rich and just under stoichiometric mixtures the foam had only a modest effect in lessening the velocity of the front. For lean mixtures, the foam has a dramatic influence in reducing DDT distances by up to 75%. This paper has some important implications for explosions in foams described by Glinka et al. (1994) and Barfuss et al. (1993), see Sect. 11.

## 10 Critical energies for initiation

Gelfand et al. (1989) confirm an earlier suggestion of mine (Nettleton 1987), based on particles serving to introduce additional turbulence into the flow, in a theoretical paper demonstrating a reduction in initiation energies for dusty mixtures.

Chou et al. (1993) and Menikov et al. (1994) have studied curved detonation waves. The first paper, an experimental one, gives measurements of critical energies required for both planar and spherical waves in mixtures of acetylene with both air and oxygen. The second one is a theoretical treatment of the effects of curvature on propagation velocities.

He (1996) using models with both one-step and reasonably detailed chemical rates for the oxidation of hydrogen shows, somewhat surprisingly, that the single reaction rate performs satisfactorily. It would be valuable to compare predictions from more complex systems, see Brown and Thomas (1999), using the optimum single-step rate, with those derived from the 600 steps they believe essential. Sochet et al. (1997) compare theoretical detonabilities, based on the total energy encompassed by the shock, with measurements for methane, propane and acetylene, claiming that there is good agreement between theory and experiment.

Achasov et al. (1995) have produced a combined theoretical and experimental study of the initiation of oxyacetylene detonations by the reflection of shocks from concave axisymmetric surfaces, from the interaction of supersonic jets of fuel and oxidant and from turbulent mixing of burnt and unburnt gases. Dorofeev et al. (1996a) have investigated the initiation of 50 $m^3$ volumes of hydrogen mixed with air by jets of combustion products exiting through nozzles of 10 to 40 cm; the minimum dimensions of these were 10 to 20 cm, in fuel concentrations ranging between 20 and 25%. As might be anticipated from work on critical diameters for the transition from planar to spherical detonation, the ratio of jet size to cell spacing is between 12 and 13. In the context of this group's work, I should draw attention to the implications for safety of their studies (Dorofeev et al. 1996b) of the amplitude of blast waves and energies radiated from fuel-rich vapour clouds.

## 11 Detonation of less common fuels

In two papers from the same group, Barfuss et al. (1993) and Glinka et al. (1994) examine detonations in foams at pressures of up to 30 bar. The first paper is an experimental investigation of bubbles both in air and oxygen of solvents, such as acetone, dodecane tetramethyl-dihydrogen-disiloxane, toluene and trichloroethylene. The second one concentrates on tetramethyl-dihydrogen-disiloxane and dodecane together with their solvents in bubbles with oxygen, producing quasi-detonations in the pressure range $5 < P < 30$ bar. It includes an empirical model predicting the influence of the partial pressure of oxygen on such fronts.

Zeitoun et al. (1995) investigated direct initiation in oxyhydrogen mixtures at 123 K, with an equivalence ratio between 0.3 and 1, and at pressures between 1 and 2 bar. At lower pressures a decrease in temperature favored the formation of the front. From the same group, Presles et al. (1996) conducted experiments on the detonative decomposition and oxidation of nitromethane at temperatures close to 390 K and pressures between 1.7 and $5 \times 10^{-2}$ bar. Both multi-headed and spinning structures were observed and, in marginally detonable media, galloping fronts. In fuel-rich mixtures all exhibited a double scale cellular structure.

Takita and Niioka (1996) have studied confined detonations in the following mixtures: hydrogen/methane/air, hydrogen/propane/air, methane/butane/air and methane/butane/oxygen, with the last mixture occasionally diluted by nitrogen. For the mixtures containing methane, velocities were those for methane only. However, the diluted methane/oxygen mixture was shown to exhibit a considerable decrease in induction time. Terao and Moyota (1992) give experimental values for C-J velocities in oxypropane mixtures over the temperature range of 400 to 700 K. Alekseev et al. (1996) examined spherical detonations in stoichiometric mixtures of gasoline with air. They worked with semi-cylinders 15 to 17 m in length and 6 to 8 m in radius (volumes of up to 1100 $m^3$). The initiation energy using oxypropane mixtures as the source was found to be 7 times that for a condensed explosive and the cell size for the gasoline detonation to be between 4 and 5 cm.

Knystautas et al. (1998) examined confined detonations in mixtures of benzene with air. They found the lower limit to be at an equivalence ratio of 0.4 and the upper 2.8 for mixtures at ambient pressure and temperature.

Gelfand et al. (1992) and Fletcher (1994) return to the earlier contention of Board et al. (1975) that explosions generated by vapors produced by the interaction of hot particles with a colder liquid are detonations.

## 12 Concluding remarks

On account of the successes of one-dimensional theory in describing the overall properties of detonations, we (*mea culpa*) have all fallen into the habit of thinking that small modifications to it can be used to treat multi-dimensional

waves. This may be a suitable time to consider its implications and to ask what we would require from an all embracing new analysis; in this particular context, of what we might like it to predict in terms of plant safety. Recall that, in these circumstances, we are likely to be dealing with either end of the composition limits and that inhomogeneities in composition are likely to be the rule rather than the exception. Typical questions posed for detonable mixtures are the parameters of the initiation source and the distance for DDT.

Answers are required as to how these are influenced by the presence or absence of confinement, by the process leading to initiation, by temperature, pressure and overall composition, and by the existence of non-uniformities in any of these features. Again, we need both peak pressures and pressure histories and how these are influenced by the confinement, particularly those occurring in structures with sufficiently short response times to deform during exposure to the wave, and to the effects of the existence of inhomogeneities in composition.

There are still other pertinent issues which remain unresolved. For instance the wide variation in regularity of structure and the patches of fine writing frequently observed when there are changes in confinement. In the former case, these are most likely to occur with the near limit mixtures of nitrogen diluted fuel-oxidant mixtures, resulting from excursions of plant. These raise queries as to whether some of the chemical and physical processes involved in ignition, particularly of dust clouds, are stochastic. Recall that the experiments by Urtiew and Oppenheim (1966) showed the various causes of the explosion within an explosion, leading to a detonation, but were unable to identify the parameters determining which was the initiating event.

Little is presently known of the influence of regularity on initiation energies and more particularly, on distances for DDT. At first sight, one might suppose that longer distances might be required for perturbations, occurring in the initiation process, to die away and produce a regular structure.

Extending the experimental studies of Edwards et al. (1981) to the propagation of single-headed rather than multi-headed fronts through bends would be valuable; not only to extend our knowledge to marginal fronts, but might produce some information on these effects. They would involve a comparison of the influence of the curvature of the bend on the fate of fronts entering with either the triple point at the outer or the inner bend. For the former re-establishment and for the latter quenching might be anticipated with changes in curvature giving an estimate of the degree of enhancement and decay at inner and outer bends respectively and their repeatability being a hint as to whether any of the processes are stochastic.

Again, we are far from understanding the effects of confinement, temperature, pressure and composition of the mixture on the regularity of an established front. A further possibility is that, when there are either rapid changes in confinement or homogeneity of the mixture, too little time is available for chemistry to approach equilibrium which may account for the formation and decay of patches of fine writing.

One of the attractions of one-dimensional theory is the empirical observation that a decrease in overall velocity of the front to 90% of the C-J value results in its extinction. This appears to hold for either thermal, viscous losses or a combination thereof and not to be greatly influenced by the composition of the mixture, though information on self-decomposing fuels is very limited. Thus, the effects of the emissivity of the reaction zones, particularly of dust flames, and of roughness of walls on both transition distances and quenching is far from being fully understood. The concept of wall roughness in this context extends from not only the finish of the surface but the inclusion of orifice plates or obstructions to promote DDT or abrupt area changes to quench established fronts. It is possible that making such demands on theories governing growth of boundary layers over such a wide range of peak widths, distributions and amplitudes are too exacting. However, even in the simplest possible case, a satisfactory explanation is lacking of the conflicting role of the surface finish of an unobstructed tube in the promotion and quenching of detonations.

At this stage, computer capacity rules out complex reaction schemes involving hundreds of reaction steps which Brown and Thomas (1999) suggest are necessary for a complete description of the oxidation of gaseous fuels other than hydrogen and possibly carbon monoxide. However, it may still be possible to treat the kinetically simpler self-decomposing fuels such as acetylene, ethylene and ozone and dusts such as aluminium and magnesium. Naturally, this situation could change with future generations of computers. Presently, it should be possible to check on the effects of non-equilibrium on simple kinetic schemes containing a slow reaction step and the influence of the properties of confinement and of mixture strength on the formation and decay of irregular triple points.

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