Some observations on the initiation and onset of detonation

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The results of experimental studies during which transition to detonation events occurred are presented. These observations and their interpretation are then discussed, and the conditions for the onset of detonation are described, with particular attention paid to the nature of the phenomena of deflagration-to-detonation transition. The resulting implications for predicting detonation evolution using computational fluid dynamic methods in practical applications are also discussed.

Keywords: gas phase; deflagration; detonation; transition

1. Introduction

Detonation and laminar deflagration represent two extreme manifestations of propagating combustion waves. Both can be represented as one-dimensional gasdynamic and thermodynamic discontinuities, consistent with the conservation laws of mass, energy and momentum, together with an exothermic energy-release term. For detonation waves, fundamental wave properties, such as supersonic wavefront velocity, temperature and pressure, may be computed accurately from basic physical and thermodynamic data. Unlike detonations, which are supersonic with respect to the reactive medium in which they propagate, deflagration waves are subsonic. In addition, although detonation relies on the close coupling of compressive shock heating and rapid exothermic chemical reaction, deflagration waves are dominated by mass and thermal diffusion processes. The propagation velocity of a one-dimensional deflagration, or the subsonic laminar flame burning velocity, is also recognized as a fundamental property of a combustible mixture, and can again be computed from a solution of the governing conservation equations coupled to the appropriate finite-rate combustion chemistry, together with models for species and thermal diffusion. A problem of much theoretical and practical interest is how a laminar flame can be transformed into a detonation. Much reliance is often placed on the use of the phenomenon of deflagration-to-detonation transition (DDT)—a term whose unqualified use may be questioned. In this paper, we present observations of instances where detonations have been observed, discuss the mechanisms by which the onset of detonation develops

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and comment on the relevance of a generalized DDT mechanism. On the basis of these observations, we further comment on the requirements for numerical simulation codes so that they may be used to predict potential practical detonation hazards.

In practice, the one-dimensional views of flames and detonations are complicated by other two- and three-dimensional effects. The gross external characteristics of typical deflagration and detonation waves are shown in figure 1, with a typical spark schlieren image of a turbulent deflagration front shown in figure 1a.

For detonation, the leading longitudinal shock wave of one-dimensional theory is, in fact, closely coupled to other transverse shock waves. For an established steady-state detonation wave, the characteristic spacing, $\lambda$, of these transverse waves exhibits well-defined dependencies on the composition, temperature and pressure of the reactive mixture. These transverse features are easily observed using schlieren techniques, as evidenced in figure 1b, or by virtue of the integrated pattern they leave on a lightly sooted surface, as shown in figure 1c. Detailed discussions of transverse structure in detonations are given by Strehlow & Engle [1] and Edwards [2]. If the detonation front is in some way forced to propagate at a faster velocity than the natural steady-state detonation velocity, then the characteristic size of the pattern decreases, reflecting the higher average temperature and pressure in this ‘overdriven wave’, as demonstrated by Thomas & Williams [3] when studying detonation interaction with inclined wedges. If, on the other hand, a wave is weakened, then the characteristic size of the detonation structure increases until, at a velocity deficit of some 10 per cent relative to the theoretical detonation velocity in that mixture, the shock and reaction separate, and the leading wavefront velocity decreases further. In one dimension, this decoupling can be engineered by lining the tube walls with an acoustically absorbent material, as shown by Guo et al. [4]. Transverse detonation structure can also be eliminated when a planar detonation wave diffracts from a pipe or duct into a larger volume, although, as will be described later, under certain critical conditions, detonation can be re-established in the resulting divergent flow field downstream of the area change.
The nature of the cellular pattern itself, e.g. characteristic dimensions and regularity, has been shown to reflect the ability of a detonation wave to propagate (see Moen et al. [5]). In general, a travelling reaction wave can be classed as a detonation if it propagates at the theoretical steady-state velocity and exhibits transverse ‘structure’. It can further be mooted that the ability to develop and sustain transverse structure is a necessary feature for detonation to exist, as it is these transverse wave interactions that promote the bulk of the energy release required in one-dimensional theoretical descriptions of a steady-state propagating detonation. Transverse structure is thus both an indicative and an essential aspect of the macroscopic propagation of a gaseous detonation wave. The existence and possible significance of transverse structure during the initiation and propagation of detonation in solid explosives have not been determined.

For gaseous mixtures or vapours in pipes, the ability of a detonation to propagate in a pipe of given diameter can be related to the characteristic size of the natural detonation transverse wave structure relative to the pipe diameter (see Dupré et al. [6,7]). Recently, following Thomas [8], (G. Thomas 2008, unpublished data) has attempted to extend this finding by seeking correlations of measured or predicted detonation cell sizes with observations of whether detonation was observed to develop in a pipe of a specified diameter. Typical initial results are given in figure 2 for a 50 mm diameter pipe. These plots present the peak pressures measured close to the end of a 1.6 m long pipe, together with measured or predicted detonation cell widths, and indicate that some form of correlation does appear to be present by which cell widths can provide a priori guidance as to possible practical concentration limits beyond which detonation will not develop in the pipe. This is indicated in figure 2 by negligible pressure development, although it is difficult to extract a definitive numerical limit criterion. It is interesting to note that the limiting critical cell width observed is significantly different from the value obtained in the earlier studies of Dupré et al. [6,7], which predict limits with cell width $\lambda$ of the order of $\pi D$, where $D$ is the pipe diameter, which is 50 mm in the studies used to obtain the data shown in figure 2. The limit data indicated in figure 2 are more consistent with the studies reported by Thomas [8] for fuel–oxygen mixtures at elevated temperatures and pressures, who found the limit to be when the critical cell size was closer to the tube diameter.
The probable explanation for the discrepancy between the two studies is that in the earlier work by Dupré, the initiation mechanism was a decaying detonation, whereas to obtain figure 2 as in the work of Thomas [8], the initiating mechanism was a low-energy spark, and so these results should be more representative of practical DDT development limits where a detonation can be expected to result after a flame-acceleration process from a low-energy ignition source.

For subsonic deflagrations, the mean propagation velocities are known to increase as transverse flow perturbations become more apparent. Turbulent variations in the flow field distort the overall flamefront, giving rise to a more convoluted surface and an increased flame surface area. Assuming that the scale of the distortions is greater than the thickness of the laminar flamefront, the overall rate of reaction (in mass terms) then increases in direct proportion to the surface area, assuming that the unburned gas density remains constant. Much of the work undertaken on gaseous explosions in recent years has been concerned with developing an understanding of the relationship between turbulent burning velocity and the intensity of the fluctuations in the unburned gas mixture flow. In the limit, following severe turbulent flame acceleration, transition to detonation occurs, as reviewed recently by Ciccarelli & Dorofeev [9]. Much effort has thus been expended over the past decades in attempting to promote this turbulence-driven route to macroscopic DDT events, often through modifying the wall roughness in tubes [10], or through the introduction of repeated obstacles. While obstacles obviously replicate some of the geometries often found in practical detonation hazard scenarios, in this author’s opinion, they can also complicate or obscure detailed observations of the final critical onset of detonation processes.

### 2. Techniques for the initiation of detonation

As mentioned earlier, a term widely used in relation to detonation is that of DDT. Often, the term is used in an inappropriate manner, and can suggest that a single process determines whether a detonation will develop, without specifying whether one is concerned with a macroscopic or microscopic process. To help illustrate the differences, some examples of means by which detonation can be engineered in reactive mixtures follow. Before proceeding further, it is necessary, to avoid possible confusion, to define exactly what is meant by macroscopic and its counterpart microscopic. When used, macroscopic will refer to dimensions comparable with a containing vessel or structure, e.g. a pipe length or obstacle dimension, while microscopic will be significantly smaller than such length scales, but greater than molecular dimensions. A microscopic volume would thus contain an extremely large number of chemically active species.

(a) Direct initiation by a high-energy source

Detonation is a unique manifestation of coupling between compressive shock heating and thermal auto-ignition of exothermic reactions. It is not surprising therefore that detonation may be initiated by direct shock effects. Planar detonations may be initiated by the reflection of a shock wave from a solid wall, as has been reported by Cohen & Larsen [11] and Gilbert & Strehlow [12], and more recently by Sands [13] and Thomas et al. [14] for shock reflection by...
discrete obstacles. Meyer & Oppenheim [15] classified the transition following shock reflection in a reactive gas mixture as either strong or weak, according to whether the detonation emerges directly from a point or surface, or is due to the coalescence of several more widely distributed ignition centres within the shocked gas volume. Knystautus & Lee [16] and Edwards et al. [17], among others, have shown that the direct initiation of spherical detonations may be effected by a sufficiently energetic point source, such as an exploding wire or chemical explosive. For energetic sources, the strength of the blast wave ensures a smooth transition from an overdriven detonation to an established steady-state detonation. Insufficient source energy results in a decaying blast wave and a trailing reaction front. Under critical conditions, a quasi-steady shock–flame complex is observed before transition to detonation occurs. In this case, the exothermic front ceases to separate from the blast, and both propagate for a short distance at an approximately constant separation until a ‘hot spot’ forms and transition to detonation results. It is seldom noted that under these conditions, detonation emerges from one or only a small number of localized regions. It is a matter of semantics, but it may thus be argued that in this case, it is not a true macroscopic DDT process, but that the blast source only creates the conditions required for the formation of hot spots, and that the ensuing final onset of the detonation phase is the true DDT process in a microscopic volume of shocked heated gas.

As the physical and thermodynamic properties of shocked gas regions differ from the undisturbed initial gas mixture, these regions are clearly defined on a macroscopic image by cellular structure that is significantly smaller than that obtained for a steady detonation in the undisturbed gas.

An example of a critical transition to detonation is shown in figure 3a, the key features of which are sketched in figure 3b. This is a reproduction of a smoked foil obtained by Thomas [18] in a sub-atmospheric acetylene–oxygen mixture using a hemispherical exploding wire blast source at the edge of a rectangular plate. The final transition leads to detonation of the shock-heated and compressed mixture. The development and propagation of detonation in this shock-heated gas can represent a significant practical hazard, as the peak pressure that is developed as the shocked gas detonates is a significant multiple of the shocked gas pressure, typical values of which were tabulated recently by Thomas [19] for onset of detonation conditions that can arise in pipeline explosion events. The transverse detonation now drives an overdriven detonation wave into the undisturbed gas, which gradually evolves into a steady-state detonation in the remaining undisturbed mixture. There are two positions slightly closer to the source, $S$, where potential transitions to detonation did not lead to sustained cellular structure, either owing to the magnitude of gradients in the flow behind the lead blast wave, or the fact that the volume of shocked gas was too small to sustain a cellular structure.

The curvature of the bands of finer structure, whose widths increase, is due to the motion of the decaying initiating blast wave and the separation of the shock and reaction front during the time it takes for a transverse detonation to propagate circumferentially in the gas volume between the leading blast and the trailing reaction zone. This feature of a band of fine detonation structure is also observed during cylindrical initiation of detonation, as shown in figure 4a, where, in this instance, two detonation onset locations can be seen, which leads to two
transverse bands of detonation in the shock-heated gas. A transverse detonation front in the shock-heated gas behind the leading blast wave is also clearly visible in the spark schlieren image of direct initiation of a spherical detonation by an exploding wire blast source placed at the apex of a solid cone shown in figure 4b. Also visible is the overdriven detonation front it causes to propagate into the undisturbed mixture surrounding the decaying blast wave.

To investigate possible critical shock strengths for the direct initiation of detonation by a shock wave, Edwards et al. [20] developed an apparatus for the direct initiation of detonation by a shock wave incident on a planar interface between inert and detonable gases. Another example of the onset of detonation that they observed, as reported by Williams [21], is presented in figure 5.

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A further experimental observation is that other transition processes can be greatly affected by additional external perturbations. Thus, for example, the critical conditions for the transmission of detonations across area changes are influenced by the proximity of reflecting surfaces, as was shown by Thomas et al. [22]. Even for unconfined spherical detonations, transition can often be identified with a small perturbation arising from the support for the flame ignition source or a weak transverse perturbation owing to wave interactions, as in the critical diffraction of a detonation into an unconfined volume. Smirnov & Tyurnikov [23] and Smirnov & Oaniflov [24] have observed several variants of the conditions that lead to the onset of the DDT process, including emergence of detonation from a flame brush, as in the case of Urtiew & Oppenheim [25,26] and Meyer et al. [27], as well as transitions owing to the shock-wave merging reported by Urtiew & Oppenheim [28].
(b) Indirect initiation

(i) Flame acceleration in a duct or pipe

An example of another sequence of events that can lead to a practical detonation hazard is illustrated in figure 6, which presents pressure histories at various locations along a 30 m long, 150 mm diameter pipe for ethylene–air and hydrogen–air mixtures, respectively, reported recently by Thomas et al. [29]. For both mixtures, an initial slow phase is apparent, followed by a rapid acceleration phase, during which a significant increase in overpressure develops. An interesting feature in the hydrogen pressure records is the significantly greater amplitude of the precursor acoustic pressure wave emanating from the initial flame-acceleration phases, linked perhaps to the larger velocity of sound in hydrogen mixtures? These eventually coalesce and lead to the formation of a shock wave, and a transition to detonation then occurs after a further short delay.

A seminal study of a flame accelerating in a duct on a laboratory scale is that reported by Laderman & Oppenheim [30], while the resulting creation of a shock wave ahead of the flame was discussed by Laderman et al. [31]. The onset of detonation following shock formation was reported in other publications by Laderman & Oppenheim [32] and Urtiew & Oppenheim [26], the former of which considered the effects of longitudinal gas-dynamic wave interactions.

At both laboratory and practical field scale, the final transition is usually attributed to a localized explosion arising from critical gradients in thermal auto-ignition delay time in the shock-heated mixture. While these and similar DDT transition processes can be explained in a qualitative manner on a macroscopic scale, accurate quantitative predictions of the initial acceleration...
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and subsequent detonation-onset process are still not readily available to process safety engineers. The more widely known commercial codes currently used for explosion development simulations seldom claim to be able to directly model transition to detonation, although their turbulent combustion modelling capabilities can give rise to conditions in the simulation that detonation experts might speculate would lead to a transition to detonation in practice. As discussed later, more precise simulation of the onset of detonation requires codes with shock formation and propagation capabilities, together with embedded auto-ignition chemistry.

(ii) Detonation diffraction

The diffraction of a detonation at an abrupt area change was first studied because it was believed to be a credible mechanism by which a donation might be established in a large unconfined external fuel–air cloud. Both laboratory and large-scale studies have demonstrated that the ease by which detonations develop or continue to propagate as a steady-state detonation emerges from a pipe or channel and diffracts into an external volume of detonable mixture is related to the relative dimensions of the pipe and the natural detonation cell size of the mixture. These experiments also showed that the same regimes could be developed as had been observed for the direct initiation of spherical detonations in unconfined volumes of detonable mixtures. As shown in figure 7a–c, these include a subcritical transition where a decaying blastwave is followed by a decoupled reaction front, a supercritical transition where detonation transverse structure develops and continues to propagate almost continuously as the wavefront transmits seamlessly into the external volume as a detonation. For critical transmission of detonation by a diffraction process, some of the original transverse structure persists along the axis of symmetry until, as shown in figure 7d–f, a transverse detonation develops in the shock-heated mixture behind the diverging blast wave, and a decoupled reaction front is generated by the action of the expansion waves generated by the diffraction gas dynamics. At the same time, an initially overdriven detonation propagates into the remaining external volume. The generation of this transverse detonation in shock-heated mixture space also develops in the numerical simulations computed by K. Vagsaether (2008, personal communication) using the FLUX LIMITER CENTRED SCHEME (FLIC) code [33], as shown in the last three frames of figure 8.

(iii) Shock–flame interaction

One practical difficulty with most studies of transition to detonation following flame acceleration is the stochastic nature of the initial turbulent flame-acceleration phase. Markstein [34], in a classic series of experiments, had shown that weak shock waves can rapidly lead to the formation of turbulent flames as they interact with an initially laminar flame. Scarinci et al. [35] extended this work, and more recently Thomas et al. [36] showed that it is possible to generate fast turbulent combustion and eventual transition to detonation in a somewhat more controlled and reproducible manner under laboratory conditions using this approach.

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Figure 7. Spark schlieren images of detonation diffraction from a rectangular channel: (a) subcritical transmission, (b) critical transmission and (c) supercritical transmission. (d–f) Spark schlieren images of critical detonation transmission as a detonation diffracts from a rectangular channel; image spacing 3 μs. Arrows indicate transverse detonation in shock-heated mixture. Reproduced with permission from Thomas [18].

Figure 9 shows a series of spark schlieren photographs for a stoichiometric ethylene–oxygen mixture diluted with 50 per cent nitrogen in a 76 × 38 mm cross-section tube at an initial pressure of 100 mm Hg. The first feature to be observed is the interaction of an incident shock wave (travelling from right to left as viewed)
with the initially oblate flame bubble. The result of this interaction is an axially symmetric torus-like flame, although the three-dimensional nature is not always well represented in two-dimensional schlieren images. The mean translation of the bubbles in the post-incident-shock flow is also evident. The incident shock velocity was $570 \text{ m s}^{-1}$. There is little significant development of any wrinkling or turbulent burning at this stage, although an acoustic pressure wave is seen to propagate upstream against the incoming post-shock flow.
At later times, the reflected wave (not shown in figure 9) can often be associated with an obvious intensification of the turbulent nature of the reaction front. In such instances, as described below, the scale of the turbulent structure changes dramatically and, as the reflected wave continues to propagate, this structure intensifies further and appears to merge with the reflected shock. This complex, termed a ‘strange wave’ by Khoklov et al. [37], continues to accelerate until eventually a detonation developed upstream of the window section. When detonation developed in this manner, the turbulent burning velocities were observed to increase from $2 \text{ m s}^{-1}$ to $ca 1000 \text{ m s}^{-1}$ in less than 0.6 ms. Under other test conditions, it would be in order to associate these reaction fronts with a turbulent deflagration but, under the given test conditions and time scales, fully developed turbulence could not be present and must develop by some other mechanism. When the test conditions are altered, e.g. a larger incident shock Mach number to that shown in figure 9, a rapid intensification of the turbulent remnants of the original laminar flame bubble occurs as the reflected wave traverses it, as shown in figure 10. The intensification of the turbulence may be aided in the test shown by the presence of the spark gap electrodes and the turbulence it generates in its wake in the incident shocked-gas flow. In figure 10, a detonation develops (second frame) at two locations adjacent to the main turbulent flame remnant and the bottom wall of the test section.

However, what relevance do these exotic laboratory-based experimental constructs have to practical safety scenarios?

One possible relevance that might be postulated is an accident scenario where a fuel leak disperses over a large area, and at the same time enters a confined volume within which it undergoes a delayed explosive combustion event, causing a jet of hot reaction products to initiate a fast deflagration in the external cloud. If, at the same time, a fragment ejected from the internal explosion struck a distant object, igniting the fuel–air cloud at that point, the ensuing remote secondary flamefront
could be perturbed by a shock front generated as a consequence of an acceleration of the initial jet-initiated deflagration. In this way, it might be possible to give rise to rapid increases in turbulent combustion rates at a significant distance from the initial explosion location. Such a combination of events might possibly have contributed, for example, to the hitherto unexplained severity of the explosion at Buncefield (as investigated by Buncefield Major Incident Investigation Board [38]) following the accidental discharge and ignition of flammable liquid petrol vapours. There appears to be much debate as to whether a detonation developed during the Buncefield incident but, as unleaded petrol mixed with air can undergo explosive ‘detonation-like’ knock events in automotive engines, it appears quite feasible to speculate that an unusual combination of precursor explosion processes could lead to local conditions that would result in a localized microscopic onset of detonation events that would eventually propagate throughout the flammable volume. Repeated obstacles are an all too obvious starting point, but other multi-stage routes to detonation can be compiled fairly readily.

3. Some earlier studies on deflagration-to-detonation transition

Several theories exist as to how transitions to detonation arise. All rely essentially on the basic theory of Zeldovich et al. [39], later represented by Lee & Moen [40] as the shock wave amplification by coherent energy release (SWACER) mechanism. Both of these mechanisms, the hot spot often invoked by experimentalists, require the existence of a critical gradient in chemical ignition delay time, which results in the onset of local reaction to be coincident with the arrival of a propagating wave. The underlying mathematics has also been discussed in detail by Kapila et al. [41], and the mechanism and conditions required for the propagation of such reaction ‘phase waves’ have also been considered by Khokhlov et al. [42] in their study of DDT in supernovae, during which they also considered the alternative possibility of generating the required conditions for detonation onset by a turbulent mixing process, together with some degree of local quenching to give the requisite spatial gradient in the progress of reaction. These authors also considered implications for unconfined terrestrial detonation.

In the work of Singh & Clarke [43], the required critical conditions arise owing to the action of a mechanical piston that leads to local gas-dynamic heating and chemical reaction. Experimentally, the generation of hot spots was demonstrated by Williams [21] (figure 5) for the case of initiation of detonation by a planar incident shock wave. Thomas et al. [44], in a follow-up study of the transmission of a detonation across an inert region, observed that onset of detonation occurred as in the earlier direct planar shock-initiation tests, but now by hot-spot formation following combustion-driven shock-front acceleration. An important point they note is that the velocity of the critical initial transmitted shock velocity was significantly less than that in the tests of Edwards et al. [20], using the same mixture. Transition was now attributed to the acceleration of the pre-existing combustion wave (decayed incident detonation reaction front) by wall-induced turbulence generated behind the weak and initially decaying transmitted shock. The final transition process however, by localized explosion, appeared to be the same in both tests, and it was postulated that the final transition to detonation process is common to all DDT events, differing only in the manner by which
the hot spot is formed. Sichel [45], in a review of the role of an ‘explosion within an explosion’ during DDT, confirmed the conclusions of Lee [46] on its significance during the final onset of detonation. Sichel also noted that turbulence is a contributing factor, in addition to other gas-dynamic processes such as shock reflection and merging, but could not identify the specific cause of the final transition to detonation.

As noted earlier, transition to detonation as a result of reflected shocks has been demonstrated by several authors, e.g. Cohen & Larsen [11], Gilbert & Strehlow [12]. Numerically, the two-dimensional effects observed experimentally by Sands [13] were predicted by Nikiforakis & Clarke [47]. A key feature in these ‘strong’ initiation cases is the fact that the ignition delay before the onset of exothermic reaction in the reactive mixtures is very short. Further, as is shown clearly by the work of Singh & Clarke [43], the time after the onset of reaction to eventual establishment of a steady detonation in strong ignition is only a small fraction of this auto-ignition delay. This is closely related to the coherence theory of the strong limit proposed by Meyer & Oppenheim [15].

Oran et al. [48] also simulated this thermal auto-ignition and onset of detonation and attempted to reproduce the experimental results presented by Cohen & Larsen [11]. For these simulations, a simplified model of ignition delay and subsequent energy-release rates were used, based on ignition delay measurements obtained from the integration of a multi-step chemical kinetic scheme at various initial temperatures and pressures. Good agreement was obtained in the case of the strong initiation of detonation, but they were not able to accurately reproduce the weak initiation results. In their simulations of a weak ignition case, transitions were only obtained by using a composition that gave a shorter auto-ignition delay than that observed in the experiments and, as a result, when transition occurred, it was manifested as a sudden transition. This is in contrast with a more extended low-velocity reaction phase observed in the experiments. This slower deflagration phase may also be compared directly with the accelerating reaction front seen in the weak ignition results of Meyer & Oppenheim [15] and Vermeer et al. [49].

4. Discussion

(a) General comments

From the brief review of DDT given in the preceding sections, one factor that appears to be clear is that while the strong ignition mechanism for detonation initiation by an auto-ignition hot spot is well understood, the weak mechanism is known only in a qualitative manner. One reason for this may be that the mixtures used previously in reflected ignition studies have, by the very nature of the experiments and apparatus, been required to have relatively short reaction times. This was done so that the gas would auto-ignite in the available test time window. The coalescence of even weak ignition centres in such mixtures is therefore very rapid in these reactive mixtures and not readily observable in detail.

Meyer & Oppenheim [15] described and discussed a coherence theory for auto-ignition and showed how the boundary between weak and strong ignition, for detonation, may be delineated by criteria related to thermal ignition delay times.

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However, Vermeer et al. [49] noted that transition to detonation also resulted from weak ignition, but that it took more time than in the strong case to ‘develop a gas-dynamic effect’. Their photographic results for i-octane show a clearly defined accelerating combustion front that leads to shock-front formation and eventual transition to detonation. For the strong ignitions in n-heptane, detonation is initiated almost immediately after the gas first ignites.

In the more recent shock–flame examples by Thomas et al. [36] outlined earlier, a reaction front seed is provided by an existing flame. Any subsequent diffusion- or mixing-controlled processes in this less-reactive mixture at intermediate temperatures then proceed, at least initially, at a slower rate and are more conducive to observation. Similar observations by Fieweger et al. [50] also show deflagration waves, which lead to DDT under appropriate conditions, although their reliance on auto-ignition leads to more stochastic onset of detonation events.

The need for an intermediate precursor turbulent combustion process, in addition to auto-ignition, in shock-heated mixture easily accounts for the inability of Oran et al. [48] to simulate the weak transition case using pure thermal chemistry, as discussed earlier. This initial deficiency was resolved in later years when new and powerful methods for adaptive mesh refinement were implemented in conjunction with monotone numerical methods. As a result, the role of intense turbulent combustion during onset of detonation simulations has now been observed in detail and is well documented [37,51,52].

As noted earlier, engineering the conditions under which a final onset of detonation event can be generated and observed under controlled initial conditions is an extremely challenging experimental task. Thomas & Bambrey [53,54], however, did manage to recreate such conditions as part of a study of the non-ideal auto-ignition using a standard reflected shock tube configuration. For certain initial conditions, auto-ignition centres coalesced to give a turbulent combustion front. Their resulting schlieren images (figure 11) show that the turbulent deflagration front undergoes a local acceleration leading to the formation of a strong acoustic pulse that might possibly steepen into a shock and which, when it interacts with a laminar flame bubble, separate from the main deflagration region, resulting in the emergence of a self-sustaining detonation in the bulk of the shock-heated mixture.

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Figure 12. Numerical simulation output showing development of conditions conducive to onset of detonation following turbulent flame acceleration in a long pipe, open at one end.

Given a deflagration or diffusion-controlled process in a pre-compressed and heated volume, the development and nature of the chemical reaction fronts are governed by the rate of energy release and acoustic dissipation times, as discussed by Bradley & Morley [55] following the calculations of Bradley [56]. At this point, it is fascinating to note that physical scale and chemical time scales apart, their descriptions are qualitatively similar to the processes observed in studies of the development of pipeline detonations, as illustrated in figures 6 and 12. First, a slow deflagration leads to an acoustic pulse. Then, after a turbulent acceleration phase, significant increases in mass reaction rates mean that the combustion energy can no longer be dissipated as easily, and a shock wave forms, leading shortly thereafter to a localized onset of detonation event, via the Zeldovitch or SWACER hot-spot mechanism in a shock-heated mixture.

From the preceding discussions, there is a strong case to postulate that, apart from strong ignition cases, the final onset of detonation, or microscopic DDT, arises owing to some final microscopic scale turbulent deflagration acceleration phase in the shock-heated mixture. The question that then arises is how the required high rates of energy release can be generated under conditions where classical turbulence cannot be contemplated and thermal auto-ignition delays are too long to give the required volumetric energy-release rates? On a macroscopic scale, e.g. for the pipeline explosion results shown in figure 6, it has proved possible using the Random Choice Method implementation described by Bjerkedvedt et al. [57], to successfully simulate shock-wave development, (figure 12), ahead of an accelerating turbulent deflagration, although the lack of a detailed finite-rate auto-ignition chemical kinetic model precluded the generation of an onset of detonation event in the simulation. The flame and energy-release acceleration rates were governed by turbulent burning rate models linked to the turbulence intensity and local mixture density in the unburned mixture ahead.
of the deflagration, where the turbulence intensity was linked empirically to the local flow Reynolds number.

A significant insight into the possible general nature of the onset of detonation phenomenon may also be gained from considering shock–flame studies and their numerical simulations by Khokhlov et al. [37]. In their initial computations, of the shock–flame experiments of Scarinci et al. [35], multiple shock reflections first lead to rapid macroscopic increases in mass combustion rates owing to baroclinic vorticity. Then, smaller amplitude pressure and flow fluctuations give rise to regions where the Zeldovich mechanism can evolve on a microscopic level in the vicinity of the reaction front. These computations, for well-resolved flamefronts, also show that at least two-dimensional simulations are required to allow the requisite increase in flame area to develop as in the experiments.

More recent three-dimensional computations, tabulated and summarized by Oran & Gamezo [52], have also shown that the intense perturbations in the flow-field parameters do not resemble classical Kolmogorov-type turbulence fields, but are highly non-equilibrium and non-isotropic in nature. These results provide the first independent evidence for the key role of chemi-acoustic interaction in the final stages of the weak mode of detonation initiation. The generation of non-equilibrium turbulence by classical mechanisms such as Richtmyer–Meshkov instabilities has been discussed by Oran [58]. By such mechanisms, the energy-release rates may not only be sufficient to become self-sustaining, but may also result in a sufficient perturbation of the unreacted gas to lead to hot-spot formation and hence detonation. This is thus an extension of the critical rates of energy release described by Bradley [56], but now via an initial deflagration combustion process, rather than a direct Arrhenius-type thermal ignition. This is consistent with the work of Meyer et al. [27], who discussed in detail the inadequacies of purely gas-dynamic process for generating the required thermal ignition delay times in their mixtures for transition to occur by the gas-dynamic route.

An example of the ability of a deflagration front to become self-sustaining without external influences has been presented by Thomas & Jones [59] for a fast, loosely coupled, shock and deflagration complex emerging into an unconfined volume, the phenomenon of the jet initiation of detonation. For the strongest emerging shock, an onset of detonation event was seen as the flame emerged, close to the surface of an external vortex of burned mixture. For the weakest incident shocks, the waves weakened owing to the lateral expansion waves generated at the area change. However, under critical conditions, the shock velocity was maintained, and this could arise only due to sufficient energy release from the trailing deflagration front, both of which propagate in an otherwise unconfined and unsupported volume. This mode of propagation is similar to that postulated by Thomas et al. [36] to explain the accelerating reflected shock–turbulent combustion complexes generated following certain shock–flame interaction events. In both cases, the mechanism by which the turbulent combustion fronts become self-sustaining or accelerate cannot be explained by existing theories and descriptions of turbulent combustion waves.

It may also be mooted that chemi-acoustic processes result in knock in internal combustion engines, when pre-compressed gas is further perturbed by intense acoustic fields that both promote the main combustion front and also act on the already compressed unburned mixture, leading to hot-spot formation. This also

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raises a further point that requires consideration when dealing with explosions in gases, that of the role of intermediate temperature chemistry.

(b) On the definition of deflagration-to-detonation transition

On the basis of the preceding experimental results and discussion, the present author—currently an advisor on industrial process safety hazards, especially potential detonation scenarios—can now come to some conclusions concerning the use of the term DDT. In particular, the term appears to be one that can be used under at least two circumstances. Firstly, there are macroscopic DDT events where an accelerating flame, or deflagration, eventually results in a propagating detonation. In the present paper, this was illustrated in figure 6 using an example of detonation development in a long confined pipe. The final transition or onset of detonation in this case is probably the result of the second manifestation of DDT, on a microscopic scale. In this case, the local onset of detonation arises owing to a hot-spot event, described by early detonation investigators as an explosion within an explosion. A common feature of this second mode for the onset of detonation is that it develops in a reactive mixture that has been heated and compressed by some precursor gas-dynamic processes; these might be either shock-wave development following flame acceleration, generation of a critical blast wave by a high-energy source, or the decay of an established detonation or adiabatic compression in an automotive engine. The final stage of this microscopic DDT event is the local onset of detonation in the shock-heated mixture because of critical gradients in ignition delay times, consistent with the Zeldovitch or SWACER mechanisms. In some theoretical circles, this final auto-ignition onset process is taken to be the definitive DDT event without giving consideration to how the hot spot was formed. However, evidence exists to suggest that its formation is in fact the result of a local non-steady deflagration event in compressed and heated reactive mixture. One might therefore describe an explosion within an explosion event as one owing to critical gradient hot-spot formation, a microscopic DDT process, where the latter is itself the result of a macroscopic DDT event.

(c) Significance of intermediate temperature chemistry

It is well known in engine combustion research circles that global activation energies decrease at temperatures below ca 1000K owing to a change in the dominant reaction-propagation mechanism. Results by Cadman et al. [60] indicate a decrease in activation energies in propane mixtures at temperatures in the region of 850–1000K and show clearly that the use of high-temperature chemical kinetics data can lead to an over prediction by nearly an order of magnitude in the ignition delay times at these temperatures. A similar study with methane was reported by Goy et al. [61], although the discrepancies observed were not as extreme as with propane. The implications of intermediate temperature chemistry are not usually widely recognized by those involved with modelling transition to detonation, although they are well known for fuels used in engines, as discussed by Bradley & Morley [55]. Reaction data for detonation modelling is however usually drawn from

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higher temperature shock tube data. Unfortunately, it is the intermediate temperature regime that prevails during incipient detonation, especially for hydrocarbon fuels.

(d) Numerical simulation and prediction of detonation development

Here, it is first necessary to reconsider the role of transverse waves in these phenomena. It was stated earlier that transverse cellular structure is essential for the propagation of a physical detonation, and this was supported by examples of the emergence of structure in observations of onset of detonation events. However, numerical modelling results presented here have shown to be perfectly capable of predicting the onset of detonation without having to model the finest details of transverse detonation structure in the shock-heated gas (see figure 11). Oran and colleagues [37,51,52,62,63], using their FLUX-CORRECTED TRANSPORT (FCT) code, have also observed rapid exothermic onset of detonation events in the shock-heated mixtures in the vicinity of turbulent flamefronts while simulating a variety of potential detonation scenarios from initially relatively low-energy ignition sources. Although they employ significant and powerful mesh-refinement techniques, it is probably unlikely that they actually resolve the fine details of the transverse detonation fronts in shock-heated gas generated during the final stages of macroscopic DDT events. One need not be surprised by this: one-dimensional models of detonation are inherently incapable of incorporating this multi-dimensional structure, yet can successfully model DDT. One must conclude therefore that, while the physical existence of transverse structure is significant and necessary for detonation to be obtained in any real physical situation, numerically, a critical requirement when modelling the onset and transition to detonation in multiple dimensions is an accurate simulation of the local rate of energy release, including the scenario-specific processes that give rise to shock-wave formation. However, once the onset of detonation occurs, the simulation code must be capable of giving rise to the development of detonation structure in the initial undisturbed gas mixture as a result of the blast wave generated by detonation onset and propagation in the shock-heated mixture, as seen in the experimental results of detonation initiation shown in figures 3 and 4. Finally, we note that in addition to local turbulent burning rates, attention should also be paid to local increases in the local mixture density immediately ahead of any accelerating turbulent combustion front caused by compression waves emanating from the combustion reaction front, as observed in the experimental and simulation outputs shown in figures 6 and 12.

The key role that can be attributed to energy release provides much opportunity for modelling DDT in practical applications. For established detonations, this is easily carried out using simple chemical thermodynamics. Predicting flame acceleration and eventual DDT is however far more problematic. For example, the results of recent studies of DDT development in pipes have shown unequivocally, as in figure 6, that in the absence of any physical perturbation, the onset of detonation is determined primarily by the extent of the flame-acceleration processes occurring immediately after ignition. Small variations in fundamental laminar flame properties can result in either transition to DDT or not, owing to their contribution to the nonlinear feedback process that can lead to shock-wave formation. These variations may be as apparently
innocuous as a few per cent change in humidity or increased specific energy-release rates if the initial mixture density increases slightly as the mixture is cooled to a few degrees Celsius, but they can lead to significant changes in flame-acceleration characteristics, mitigating or promoting DDT. One of the major challenges to the modelling of DDT in this configuration is thus the development and successful application of transient turbulent flow models in pipes and the associated turbulent combustion models. However, in addition to accurately modelling turbulent combustion, practical detonation explosion hazard simulation codes must also have the intrinsic capability to resolve and capture shock-wave formation, together with a capability for the inclusion of finite-rate auto-ignition chemistry.

5. Concluding remarks

While it is now universally accepted that it is the hot spot or Zeldovich mechanism that is the cause of the ultimate transition to detonation process in reactive mixtures, the ability to predict the onset of detonation in an \textit{a priori} manner for a set of arbitrary initial conditions must take account of the processes that lead to the formation of the required critical gradients. Strong or direct initiation may be easily achieved and predicted for strong shocks and reactive mixtures, i.e. mixtures with relatively short auto-ignition delay times. For less-reactive mixtures, and less-energetic ignition sources, macroscopic turbulent combustion and flame-acceleration models with intrinsic shock-wave capture capabilities, as well as intermediate microscopic descriptions of turbulent deflagrations in shock-heated mixtures are required, the latter corresponding to that defined by early investigators as weak ignition.

In general, one can summarize the available paths that can lead to detonation via the different modes sketched in figure 13. For high-energy sources, such as solid explosives, detonation develops continuously as the overdriven blast wave...
decays asymptotically to the steady-state detonation velocity. As the source energy is decreased, a shock with a trailing deflagration in the now shock-heated mixture develops. The onset of detonation occurs in the vicinity of the trailing turbulent flamefront, presumably in a manner similar to that shown in figure 11. A similar shock–flame complex develops for low-energy ignition sources if some mechanism exists to promote an initial rapid flame acceleration. As soon as a shock wave forms, the onset of detonation can now evolve in the the shock-heated mixture. Given the experimental evidence, one might speculate that the development of a shock–turbulent deflagration complex in the shock-heated mixture is a universal and necessary precursor to any final onset of detonation event for all mechanisms for generating detonations, other than that of supercritical direct initiation by a high-energy initiation source.

The phenomenon of flame acceleration in a confined volume due to turbulence is well known, and can be assumed to be responsible for the initial stage of slow deflagration on a macroscopic scale. However, as the gas-dynamic conditions and time scales change, it is argued that continued explosion development is now due to the maintenance and amplification of combustion by intense acoustic interactions on a microscopic scale. These interactions then give rise to reaction fronts that, in outward appearance, resemble classical turbulent pre-mixed flamefronts but which, in reality, are supported by non-equilibrium non-isotropic turbulence. In the limit, the three-dimensional pressure fluctuations that are generated lead to the formation of the transition hot spot in pockets of unburned mixture or volumes of mixtures that will be sensitive to additional perturbations, such as weak shock waves. This phenomenon has been shown to be significant in detonation-initiation studies in the laboratory, but it is also likely that it is the dominant mechanism in the final transition process under a far wider range of initial conditions. It may similarly lead to knock in internal combustion engines.

It would seem therefore that one of the key challenges remaining to complete our understanding of the ultimate onset of a transition to detonation process is the development of a better understanding of the generation and characterization of intense non-equilibrium non-isotropic turbulence and its interaction with exothermic reaction fronts. The reader should note that these phenomena differ significantly from the classical description of turbulence-based DDT, which are macroscopic processes dominated by obstacles or pipe walls interacting with the bulk flow. For large fuel–air clouds, an early study of how detonation may develop was reported by Moen et al. [64], in which turbulence generation owing to the presence of obstacles and the concurrent influence of confinement was clearly illustrated. This large-scale study built on initial laboratory studies of the influence of repeated obstacles in generating a positive flame-acceleration feedback loop via turbulence generation by obstacles [65,66].

Large-scale DDT events promoted by obstacles rely on a macroscopic scale energy cascade through progressively smaller length scales, via the usual Kolmogorov or Richardson mechanisms that eventually perturb the chemical energy-release mechanisms. The turbulent-like processes pertinent to microscopic onset of detonation, as visualized in figure 10, are however not understood and appear to offer a new and exciting area of study for turbulent combustion specialists. One starting point might be that of fractal-generated turbulence, as described recently by Mazellier & Vassilicos [67], but where the fractal energy
source perturbing the flow might be a turbulent combustion front as opposed to a solid fractal grid. This situation might then possibly provide the basis for a microscopic combustion acceleration feedback mechanism in reactive mixtures, especially if the mixtures were compressed and heated.

In addition, for explosions, as in engines, the role of intermediate temperature chemistry requires careful scrutiny.

Finally, while the preceding discussions have been concerned with gas-phase detonation phenomena, experimental evidence presented by Bernecker & Price [68–70], Gipson et al. [71] and Luebcke et al. [72] displays features reminiscent of those developed in the experimental results shown in figure 6 and the corresponding simulation output given in figure 12. This would suggest that the microscopic mechanism of rapid combustion front acceleration and precursor shock formation is one that is obtained during DDT events in solid explosives as well as detonable gaseous mixtures. In the studies with solid explosives, we note the emphasis placed on the density changes ahead of the reaction front because of the compression waves emanating from the reaction zone.

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