On the Diverging Spherical Detonation and the Critical Tube Diameter

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1 Brief History of Spherical Detonations

The major interest of spherical diverging detonations concerns the conditions of existence of detonations. Critical conditions of their initiation, either by point source ignition or by a detonation diffraction from a tube into free space, provide a good estimate of the detonability of a reactive mixture, via the notions of the critical initiation energy and the critical tube diameter of transmission of a detonation, respectively.

The absence of confinement renders the problem intrinsically “clean”. Only divergence competes with the chemical energy release.

After the formulation of his hydrodynamic theory of detonation, Jouguet [1], in 1917, did not believe that the self-sustained diverging spherical detonation can propagate at a constant velocity independent of the hydrodynamic field behind it, i.e.:

« rien ne dit précisément que ce mouvement restera ce qu’il faut pour que la célérité soit constante »

Nevertheless, Lafitte [2] in 1923 succeeded in observing spherical detonation having the same velocity as plane detonation in $CS_2 + 3O_2$ and in $2H_2 + O_2$ mixtures. As these waves were initiated by relatively powerful detonators (1g of mercury fulminate) and recorded over rather short distance (10-13 cm) some doubt raised on their true nature.

During the second world war, Taylor [3] on the one hand, and Zeldovich [4] on the other, examined solutions capable of representing the movement of the burned gas behind the combustion wave compatible with the CJ condition. A self-similar solution for the detonation field produced by diverging detonation wave was then expressed assuming the existence of infinite gradients just behind the detonation front of zero thickness.

N. Manson, in 1954, with his colleague Ferrié [5] undertook an experimental investigation of the spherical detonation in order to verify if the aforementioned theory is valid. He knew the work of Lafitte with the inherent caution about the autonomy of the detonation wave already observed. Experimental results obtained in larger volumes than those used by Lafitte and with different point ignition sources confirmed the previous theory i.e., the spherical detonation velocity was very close to the CJ value for different reactive mixtures (C_2H_2/O_2, C_3H_8/O_2, CH_4-C_2H_4/O_2, St Marçay’s natural gas/O_2).

Later, co-workers of N. Manson, i.e. J. Brossard and R. Cheret, addressed again the diverging spherical and cylindrical detonation propagation in gaseous and in condensed matter in order to obtain some new detonation insights as (i) wave structure, (ii) asymptotic detonation velocity (when the detonation wave radius tends to infinity) [6] and (iii) some wave curvature-velocity relationship [7]. In 1981, Desbordes et al. [8] measured accurately the pressure profile inside the quasi CJ self-sustained spherical detonation wave ignited by a point source and confirmed the Taylor-Zeldovich theory thus eliminating the controversial weak-detonation solution in fuel-oxygen mixtures (see Fickett and Davis [9]).

At the same period, a large international effort has been directed on the problem of the direct initiation of spherical detonation in order to understand the real physics (Litchfield et al. [10], Bach et al. [11], Struck [12], Lee et al. [13]). Some phenomenological models were then built aiming at describing the main mechanisms involved (Chernyi and Levin [14], Lee [15]).

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The pioneer work on the key issue of direct initiation of detonation (by point blast and planar initiation) is that of Zeldovich et al. [16] in 1956. For the first time, a criterion was established on the dependence of the critical initiation energy E_c to the chemical induction length L_i, i.e.:

$$E_c \sim L_i^3$$

for spherical diverging shock wave decaying to the CJ detonation wave pressure level. This expresses the competition between the expansion behind the spherical decaying shock wave and the chemical energy release behind it. In essence, the energy source would be able to create a spherical shock wave of sufficient strength ($p \sim p_{CJ}$) over a minimum thickness (L_i) in such a way that chemical reaction couples to the shock wave turning into a CJ spherical detonation. This criterion is only qualitative but of great fundamental significance linking the hydrodynamics and the chemical kinetics of the initiation process.

The second result of the same work underlines the previous result and was perhaps at the origin of the above mentioned criterion. Quantitative data on critical diameter of transmission of a detonation from a tube of diameter d into free space established, for eight different reactive mixtures, another critical initiation criterion, viz.,

$$d_c = 15\ L_i$$

This work, after the discovery of the cellular structure of detonation and the link postulated by Schelkhin and Troshin [17] between its characteristic size λ (its width) and the chemical induction length L_i, was followed by that of Mitrofanov and Soloukhin [18] in 1964 for the C_2H_2/O_2 system where correlations were found between the critical diameter and the cell size:

$$d_c = 13\ \lambda$$

for tube of circular section, and

$$d_c = 10\ \lambda$$

for tube of square section.

These fundamental findings were followed by a lot of studies, particularly by the work of Matsui and Lee [19] who explored the detonability of many different fuel/O_2 and C_2H_2/air systems. Edwards et al. [20] showed some discrepancies with the previous correlation for systems presenting very instable cellular structure. Nevertheless, a clear failure of the $d_c=13\lambda$ correlation was shown by Moen et al.[21] for C_2H_2/O_2 mixture highly diluted by argon and later by Desbordes et al.[22] in mixtures highly diluted (more than 50% in volume)by monatomic gases as helium, argon and krypton. For more than 80% dilution, the critical tube diameter becomes unquestionably d_c=26-30 λ.

During this time, the Zeldovich et al. criterion was revisited in the light on the one hand of the point blast wave theory and, on the other hand, of the planar detonation diffraction. A few semi-empirical theories gave a better quantitative approach to the critical initiation energy of diverging spherical detonation (see Benedick et al. [23]), E_c being generally expressed by:

$$E_c = A\gamma p_0 D_{CJ}^2\, \lambda^3$$

At critical conditions, experiments showed (Desbordes [24]) that a quasi spherical pre-detonation region of mean critical radius R_c exists in both situations (point blast or plane detonation diffraction initiation) linked with cell size by the relationship:

$$R_c = 20\ \lambda = 1.6\ d_c$$

This result could be compared to that of Murray and Lee [25] who determined the critical condition of detonation front curvature i.e., R_c = 14-15 λ, necessary for its propagation in a slab geometry with lateral expansion of the detonation products.

Attempt of rigorous theoretical models was made since the ninety's. In the theoretical model of He and Clavin [26] based on quasi-steady solution of curved detonation, with the assumption of large R/L_i, it was shown that a sonic solution at the rear of the reaction zone is only obtained for a minimum value of $R/L_i=R_c/L_i$ ranging from 300 and 700, depending on the chemical system used. The non linear curvature effect of the detonation front was then recognized as the mechanism of detonation initiation failure.

Few years later, for Eckett et al. [27] the primary failure mechanism of point blast wave initiation of detonation was not the wave front curvature but the unsteadiness in the induction zone arising from the deceleration of wave front.

Both works found the expression of critical initiation energy as previously written in the above phenomenological models using the point blast wave model. Nevertheless, in these models three-dimensional unsteady cellular aspects of detonation are ignored.

At the same time, numerical simulation of cellular detonation propagation and of initiation of detonation by diffraction of a planar detonation from a tube of circular section to free space without and with obstacle in the tube exit provide semi-quantitative very good results on critical initiation conditions (cf Khasainov et al. [28]).

In this paper, based on results of experiments and numerical simulations, we present new insight into spherical detonation initiation by plane detonation diffraction. Based on results obtained with two chemical reaction-step systems as H_2/NO_2 mixtures [29] showing very fast first exothermic reaction followed by a much slower one, we present an attempt to justify the large discrepancies of critical diameter correlation for initiation of spherical detonation in reactive mixture highly diluted by a monatomic gas if compared with the same undiluted mixture.

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2 The critical tube diameter

As explained above there are many gaseous reactive mixtures where the critical tube diameter of transmission in free space obeys more or less the d_c=13 λ rule. They are essentially classical fuel-oxygen mixtures close to stoechiometry, eventually with a limited amount of diluent. Correlations for very rich and lean fuel/O_2 mixtures which have not been explored systematically are really not known. Few experiments in stoechiometric fuel/O_2/N_2 have been performed in the past and up to the air as oxidizer for a limited number of fuels (the more detonable as C_2H_2, H_2 and C_2H_4). In general, with air, the cellular structure is very irregular with substructures inside and therefore it is difficult to measure accurately its cell size and consequently to obtain good critical tube diameter correlation. Indeed, for H_2/air stoechiometric mixture at ambient conditions, the correlation first announced as 13λ [30] now is admitted as 18-24λ [31]. Some tests performed with CH_4 and C_3H_8 in oxygen and nitrogen mixtures result in d_c/λ of 24 and 21 respectively (cf [32]).

In a recent study presented elsewhere [29], we addressed the detonation diffraction of H_2/NO_2 mixture with two-steps energy release. In that mixture, the total heat Q is released in two successive exothermic steps of respective energy Q_1 and Q_2. An example of the ZND reaction zone structure for ϕ=0.5 is displayed Fig 1. The characteristic chemical time of the first step (corresponding to the delay of the maximum heat release rate behind the leading shock wave) is very short, typically 10^{-7}s in standard conditions (p_0=1bar, T_0=293K) and depends weakly on equivalence ratio ϕ, in the range 0.5< ϕ <1.3. Associated to this time is the cell net of small size λ_1. Depending on ϕ, a second maximum of reaction rate (i), exists for ϕ >1 typically one to two order of magnitude later than the first one and, (ii) does not exist for ϕ <1. It was shown experimentally and numerically that the existence of a second maximum of reaction rate is at the origin of the larger cell net in the double-cell systems [33, 34].

Detonation diffraction in such systems exhibits non-classical behaviour, i.e.:

- in single-cell detonation ($\lambda = \lambda_1$) corresponding to ϕ < 1, the critical tube diameter for successful transmission of the detonation must contain more than 13 cells: d_c/λ_1 is an increasing function of ϕ close to 17-18 for ϕ = 0.5 and equals to 45-50 for ϕ ~ 1. A so large discrepancy from the classical d_c = 13 λ law is due to the fact that λ_1 is only representative of the chemical kinetics corresponding to the Q_1 part of the whole chemical energy release Q. $K = Q_1/Q$ is a decreasing function of ϕ equal to approximately 2/3 for $\phi = 0.5$ and 1/3 for $\phi \sim 1$.

- in double-cell detonation corresponding to $\phi > 1$, d_c/λ_1 continues to increase with ϕ varying from 45-50 for $\phi \sim 1$ to about 100 for $\phi = 1.3$ while the ratio K remains constant of about 1/3, independently of ϕ. The critical diameter can be also established considering the cell size λ_2, for which Q_2 represents 2Q/3. A quasi-constant relationship $d_c/\lambda_2 \sim 3.5\text{-}4$ is then shown.

Some analogy can be found if we examine detonation diffraction in H_2/NO_2 lean mixtures and in a reactive fuel/O_2 mixture diluted by a large amount of argon or other monatomic diluent. Increasing argon dilution changes progressively the energy release law. For instance, examining the steady ZND reaction zone in $C_2H_2+2.5O_2+ZAr$ mixtures as Z increases from 0 to 15 (81% Ar dilution), we notice that beyond Z=3.5 (50% Ar dilution), a second very slow heat release step appears clearly behind the first very short step, see Fig 2 [35]. The first step is associated mainly to oxidation of C into CO and the second to oxidation of CO into CO_2. No delay can be associated to the second step of duration typically two orders of magnitude larger than the first one, and accordingly no second-cell net appears. So, the unique cell observed represents only the first part of the energy release, as for H_2/NO_2 lean mixtures for $\phi > 0.5$.

For dilution by 81% argon, d_c equals 26-30λ, as in H_2/NO_2 mixture for $\phi = .8$. In both mixtures, observation of critical tube diffraction of the detonation reveals that cells submitted to the head of the expansion fan are not destroyed but enlarge before their progressive failure. In other words, the coupling between shock and reaction front is maintained if submitted to 2D expansion. Once the structure is destroyed by further expansion, ignition of the transverse super detonation happens in the post shock reactive mixture at a finite distance from the tube exit where curvature becomes moderate. In the case of a one-step exothermic reactive mixture, cells are, in general, quasi instantaneously destroyed (decoupling appears instantaneously) as soon as detonation is submitted to the head of the expansion fan limiting the unperturbed cone (see Vasiliev [36], Lee [37] and Pintgen and Shepherd [38]).

Probably, in the above mixtures considered, during the diffraction, the second reaction step of long characteristic time is completely destroyed and is therefore not operational for the detonation re-initiation and propagation at short distance from the diffraction. This can be compared to what happens in double-cells detonation systems when "low velocity detonations" are observed in tubes because the second reaction step is no more included in the detonation reaction zone if limiting conditions are attained (see [39] and [40]). Then detonation propagation supported only by its first step once submitted to expansion, cell of larger size representing really the first reaction step alone emerges before to be destroyed progressively. In that case, using the classical hydrodynamic theory, a loss of 20% of the chemical energy for instance, (i) reduces the detonation velocity by 10% about and (ii), for systems of low activation energy (i.e. $Ea/RT_{ZND} \sim 5$ as for large argon dilution), increases the chemical time and cell size by 100%. So, successful detonation diffraction in a two-step reaction mixture needs a number of cells in quasi CJ regime, representing only a part (the first) of the total energy release, larger than in classical one-step reaction systems.

Another observation that demonstrates the existence of a second exothermic reaction step of long duration in highly argon diluted reactive mixtures can be deduced from a very simple experiment. A long run self-sustained quasi-CJ detonation propagating in a circular tube in $C_2H_2+2.5O_2+15Ar$ mixture and containing in the diameter of the tube about 10 cells or more, encounters a slightly diverging cone of 5° half summit angle. As clearly shown in [31], the detonation entering the cone slows down very progressively, its velocity losing a tenth of percent while the cell size increases twice and a bit more continuously before to stabilize. If the cone is long enough and for a large number of cells included in the diameter of the tube, it is possible to observe the detonation reacceleration towards the quasi-CJ regime as its cell size decreases. The same experiment with $C_2H_2+2.5O_2$ mixture differs markedly from the previous showing that detonation cell size and velocity are unaffected by the change of geometry for 5-7 cells detonation in the diameter of the tube.

Thus, cellular structure of such two-steps systems of self-sustained propagation regime in tubes (i.e. quasi CJ wave) can be thought as that of a one-step (the first) detonation wave pushed by the energy release by the second step. In such case the cellular structure is rather regular. Suppressing the second step of the self sustained detonation by a small divergence results in the emergence of irregular cells with a substructure inside. For this low velocity detonation regime, the critical tube diameter should be very close to the classical $d_c=13\,\lambda$ rule.

Guilly et al.[41] performed recently a numerical simulation of the detonation diffraction in C_2H_2+2.5O_2+15Ar mixture, approximating accurately the heat release law of Fig 2 by two successive Arrhenius steps. The result shows that critical tube diameter is d_c =32λ while experiment gives d_c ~ 26-30λ. The same calculation, based on one Arrhenius step for C_2H_2+2.5O_2 mixture, validates the d_c ~ 12λ correlation obtained experimentally [28].

3 Conclusions

Examining the ZND detonation reaction zone of C_2H_2+2.5O_2+ZAr mixtures for large Z , it has been shown that chemical energy release law differs from that of the undiluted mixture. It is in particular characterised by the occurrence after the first ever-present short delay exothermic step of a second exothermic step of very long duration (without any maximum in the heat release rate). The first step producing a maximum heat release rate is always responsible of the cellular structure of the detonation.

This non-monotonous heat release law controls the detonation behaviour once it is submitted to limited or abrupt expansion. The larger is the relative amplitude of the second energy step, the larger are the number of cells (of the first step) needed for the critical transmission from a tube to free space. This is also observed in H_2/NO_2 lean mixtures where energy release occurs in two steps.

This leads one to the conclusion that the cell regularity in high monatomic gases diluted mixtures is not responsible of the breakdown of the d_c=13λ correlation. Thus, the schematisation of the energy release in detonation reaction zone by a global one step of Arrhenius form represents poorly what happens really in different chemical systems. The chemical heat release law is more complex and can change during the transition from two-step to one-step only mechanism.

All these features highlight that the conditions of existence of detonation in free space and its behaviour in other given hydrodynamic configurations is intimately governed by the energy release law via its own detailed chemical kinetics, and that detonation dynamic parameter correlations established only with the detonation cell size give an incomplete view of the complexity of its dynamics. Probably, for a one-step energy release detonation system, the d_c=13λ law may be universal, otherwise for other systems as presented above the number of cells in the tube diameter needed for spherical detonation initiation must be larger.

4 Dedication

This paper is dedicated to Numa Manson, my master. I have known Numa since the last 60’s when student I undertook a thesis under his supervision on the subject of spherical detonation. During my scientific formation, I remember the few sabbatical visits of Tony Oppenheim and Rem Soloukhin in Poitiers and how I follows with interest their lectures. I remember, too, many exciting, tremendous, controversial and in fine lively discussions on detonation between the three “fathers” of ICDERS.

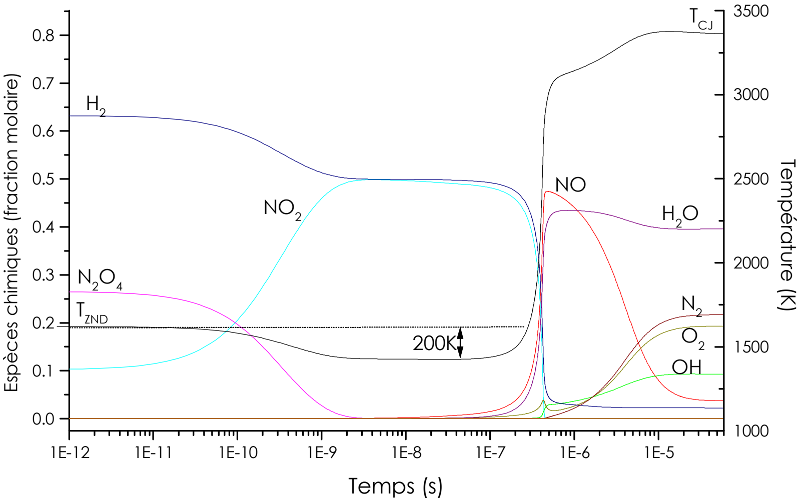

Fig 1: *Time evolution of temperature and main chemical species in the ZND reaction zone of H_2-NO_2/N_2O_4 mixture for ϕ=0.5, p_0=1 bar, T_0=293 K.*

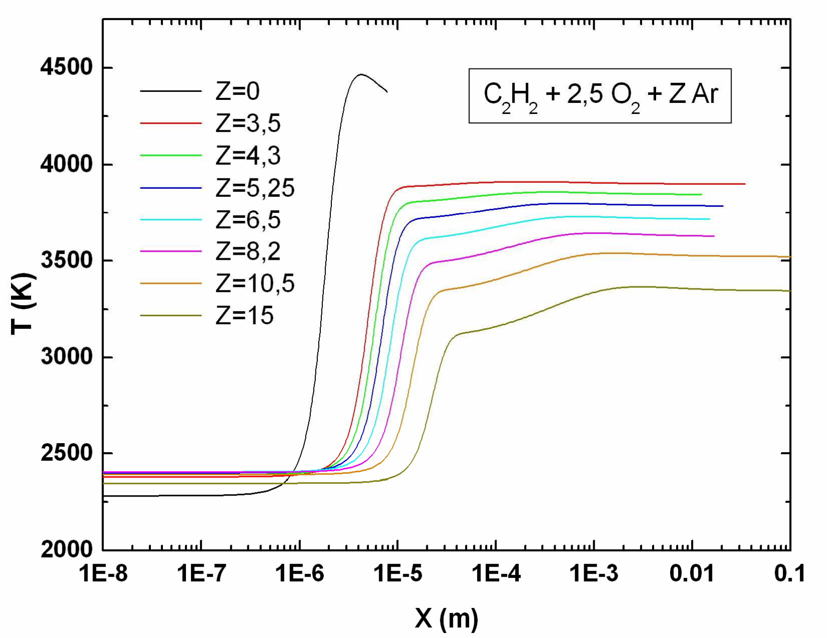

Fig 2: *Temperature evolution in the ZND reaction zone of C_2H_2 +2.5O_2 + ZAr mixture for p_0=1 bar, T_0=293 K, in function of Z (0<Z<15).*

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