Existence of the detonation cellular structure in two-phase hybrid mixtures

B. Veyssiere, W. Ingignoli

Laboratoire de Combustion et de Détonique, UPR 9028CNRS, 1 avenue Clément Ader, BP 40109, 86961 Futuroscope-Chasseneuil, France

Received 10 May 2001 / Accepted 12 August 2002 Published online 19 December 2002 – © Springer-Verlag 2002

Abstract. The cellular detonation structure has been recorded for hybrid hydrogen/air/aluminium mixtures on $1.0 \text{ m} \times 0.110 \text{ m}$ soot plates. Addition of aluminium particles to the gaseous mixture changes its detonation velocity. For very fine particles and flakes, the detonation velocity is augmented and, in the same time, the cell width λ diminishes as compared with the characteristic cell size λ_0 of the mixture without particles. On the contrary, for large particles, the detonation velocity decreases and the cell size becomes larger than λ_0 . It appears that the correlation law between the cell size and the detonation velocity in the hybrid mixture is similar to the correlation between the cell size and the rate of detonation overdrive displayed for homogeneous gaseous mixtures. Moreover, this correlation law remains valid in hybrid mixtures for detonation velocities smaller than the value D_{CJ} of the mixture without particles.

Key words: Detonation, Cellular structure, Two-phase mixtures, Hybrid mixtures, Aluminium

1 Some features of the detonation cellular structure in gaseous and solid particle-gas mixtures

1.1 Gaseous mixtures

Since the work of Denisov and Troshin (1960), a large amount of experimental as well as numerical work has been devoted to the study of the so-called cellular structure of detonations. This aspect of detonations has been extensively investigated in gaseous mixtures, which has displayed the importance of studying this threedimensional cellular structure for the understanding of propagation mechanisms of the detonation wave. It is now recognized in gaseous mixtures that this particular structure may be considered as a "signature" of the detonation. It permits to characterize the formation, the steady propagation and the extinction of the detonation regime. Moreover, the size of the elementary cell depends on the actual composition of the gaseous mixture and on initial experimental conditions. The characteristic parameter used is the cell width λ . It has been shown that the value of λ is related to the mean chemical induction length; thus the knowledge of λ provides information about the detonability of a gaseous mixture. According to this idea, extensive experimental work has been achieved by numerous authors for determining the characteristic cell size λ

Correspondence to: B. Veyssiere (e-mail: veyssiere@lcd.ensma.fr)

tial conditions. Furthermore, correlations between the cell width and certain characteristic dimensions of the surrounding confinement in which the detonation propagates have been derived which permit to build up criteria for critical diameter of propagation of a detonation, critical initiation conditions, critical conditions for transmission from a tube to unconfined or semi-confined medium, etc. Of particular interest are the results of Desbordes (1988) showing in the case of strong detonations the dependence of cell width λ on the current value of the detonation velocity D. He showed that the value of λ was not only a characteristic parameter of the self-sustained steady Chapman-Jouguet detonation, but of any strong detonation wave: when the detonation velocity D is increased above the value $D_{\rm CJ}$ of the Chapman-Jouguet detonation, the cell width diminishes, which means that the induction length of chemical reactions behind the incident shock wave diminishes correspondingly. He showed that the correlation between the variations of cell width λ , induction length L and detonation velocity D could be predicted analytically by the relationship (1):

of gaseous mixtures under various composition and ini-

$$\frac{\lambda}{\lambda_{\rm CJ}} = \frac{L_{\rm ind}}{L_{\rm ind, CJ}} = \frac{D}{D_{\rm CJ}} e^{\frac{E_a}{RT_{\rm ZND}} \left\lfloor \left(\frac{D_{\rm CJ}}{D}\right)^2 - 1 \right\rfloor}$$
(1)

where λ and $\lambda_{\rm CJ}$ are the cell widths, $L_{\rm ind}$ and $L_{\rm ind,CJ}$ the global chemical induction lengths, D and $D_{\rm CJ}$ the detonation velocities, respectively of the strong and Chapman-Jouguet (CJ) detonations. It is worthy to recall that



Fig. 1. Variation of the detonation cell width with the Mach number of the detonation wavefront for acetylene-oxygen mixtures diluted with argon (i = 0; 1; 3.5) – from Desbordes, 1988 – (with courtesy of D.Desbordes)

Eq. (1) is based on the dependence of the chemical induction length on the leading shock strength (which is characterized by the velocity D) and does not include any assumption about the stationarity of the detonation, nor on the relative values of D and $D_{\rm CJ}$ (Eq. (1) is analytically valid for any value of D). Illustration of this λ -D dependence taken from results of Desbordes (1988) is given in Fig.1, in the case of detonation of acetylene-oxygenargon mixtures with various rates of overdrive above the Chapman Jouguet velocity (M_S is the Mach number of the overdriven detonation and M_{CJ} that of the Chapman-Jouguet detonation, thus $M_S / M_{\rm CJ}$ measures the rate of overdrive of the detonation. Obviously, $M_S / M_{\rm CJ}$ is physically bounded to values greater than 1 in experiments).

As demonstrated by Desbordes, the cell size is extremely sensitive to the rate of overdrive and varies exponentially. This exponential variation derives from the dependence of the induction length on the temperature behind the leading shock front. Hence, in his case a small rate of overdrive of the detonation results in a drastic change in the detonation cell size: this is clearly observed in Fig. 1 where the cell size is divided by two for a rate of overdrive of only 1.07.

1.2 Two-phase heterogeneous mixtures (solid particles)

Two phase heterogeneous mixtures with solid particles are defined as mixtures in which the gaseous phase contains only the oxidizer, whereas the combustible component is in the solid phase, well distributed in small solid particles in suspension in the gaseous phase. Even if at macroscopic level (global heat release), one can consider that a certain similarity exists with the detonation of premixed gaseous mixtures, it is obvious that the kinetics of heat release between combustible and oxidizer in two-phase mixtures strongly differs from that of homogeneous gaseous mixtures (due to thermomechanical interphase exchanges).

Existence of truly self-sustained detonations in such two-phase media vet remains a not completely clarified problem. First experiments have been performed by Strauss (1968), who displayed detonation regimes in aluminium oxygen suspensions contained in 26.4 mm- and 44 mm-diameter, 2.7 m-long tubes, and initiated with strong sources. The observation of spinning detonations indicates that the propagation regime evidently depended on the confinement and probably was not self-sustained. Available experimental results on this problem are in limited number and in most cases no irrefutable conclusion can be drawn from them, because the diameter of the confinement and the distance of propagation of the detonation wave were insufficient to provide the guarantee that the detonation was truly self-sustained. This is the case of experiments of Kaufman et al. (1984), Peraldi and Veyssiere (1986), Wolanski (1991), Li et al. (1993) and Borisov et al. (1991). More conclusive are the works of Zhang and Grönig (1991, 1993) and Zhang et al. (1992) on the study of detonation in cornstarch and anthraquinone particles dispersed in oxygen and air. As for experiments of Gardner et al. (1986) they have been performed in a tube of diameter significantly larger (0.6 m) than those of above studies; however the length was too short to ensure that a self-sustained detonation had been observed at the end of propagation. Moreover, in their experiments, analysis of the phenomena is more complicated, due to the use of coal dust: indeed, heat release supporting detonation propagation may come from both volatile and solid components of the coal particles; such a situation rather corresponds to that of hybrid mixtures which will be examined in Sect. 1.3. Initiation of a detonation has been attempted by Tulis and Selman (1982) in unconfined cylindrical aluminium-air clouds, but their results are not conclusive. Ingignoli et al. (1999a) have tried to perform direct initiation of a detonation in hemispherical unconfined $(0.4 \,\mathrm{m}^3)$ clouds of aluminium particles in pure oxygen. Their experiments, as well as numerical simulations display that the volume of the cloud should be larger, at least by four times, to expect observation of a self-sustained detonation. Recent works in tubes have provided new information. Pu et al. (1997) have observed, at the end of 0.14 m diameter, 12 m long tubes filled with aluminium dust-air suspensions, the propagation of quasisteady propagation regimes with typical velocities of the order of 2000 m/s, that is higher than the value of the detonation velocity derived from the thermodynamical theory. The recent work of Zhang et al. (2001) utilized two detonation tubes 0.14 m and 0.3 m in diameter with a lengthdiameter ratio of 124. They observed DDT to a detonation governed by the existence of transverse waves in cornstarch, anthraquinone and aluminium particles suspended in air. Even in this case, the detonation wave is typical of spinning detonation regime and a relatively strong initiation source is required when compared with gaseous DDT.

Referring to the existing knowledge in gaseous mixtures, it appears of great importance to search whether the detonation regime in two-phase mixtures may exhibit the so-called cellular structure. However, nothing much is known about the process through which heat release from reactions of solid particles with a gaseous oxidizer can support detonation propagation. The characteristic time of heterogeneous reactions between particles and gases is generally far larger than that of homogeneous gaseous reactions by an order of magnitude or more, depending on particle size. Thus, the coupling between the shock front and the reaction zone is believed to be weaker than that existing in gaseous detonations. However, should the fundamental mechanisms of coupling between the shock front and the reaction zone be of the same nature as for the gaseous mixtures, the detonation cellular structure should exist. According to the difficulty to generate detonations in two-phase mixtures and to the larger values of characteristic time of reactions between particles and gases, the characteristic width of the cellular structure should be greater by an order of magnitude or more than for gaseous mixtures.

Until now, proofs of existence of the cellular structure in two-phase mixtures are extremely limited: In their experiments in unconfined clouds of aluminium particles suspended in oxygen, Ingignoli et al. (1999a) have recorded a few cellular-like structures with a characteristic dimension of $5-10 \,\mathrm{cm}$. But these observations have been done at the external boundary of the cloud, thus it cannot be concluded that these structures would exist at a further stage of propagation. Zhang et al. (2001) have reported to have observed the cellular structure in cornstarch-oxygen mixtures at 0.5 bar initial pressure: on smoked-foil disposed at the walls of a 0.3 m diameter tube, they have registered between one and two cells within the tube circumference. These observations are corroborated by pressure registrations made with seven pressure transducers located around the circumference of a cross section of the tube. The average value of the cell width λ obtained by these two techniques is, under their experimental conditions, of the order of 0.50 m. With aluminium-air mixtures at 1 bar initial pressure, only results obtained with the multiple pressure transducers technique are reported and indicate a cell size of about 0.4 m. It is worthwhile noticing that the cell size of dust detonations strongly depends on the particle size and shape.

1.3 Hybrid mixtures

Hybrid mixtures differ from two phase heterogeneous mixtures by the feature that the combustible is provided both by the solid particles and the gaseous mixture. This results in the existence of two different characteristic times in the heat release process, since the characteristic time of reactions between particles and gases strongly differ from that of homogeneous reactions between gaseous components. Existence of truly self-sustained detonations in hybrid mixtures has been displayed only in a few cases (Veyssiere 1986). To acquire a better understanding of mechanisms of detonation propagation in such complicated systems, specific investigations have been conducted in hybrid mixtures made of detonable gaseous mixtures with suspended reactive solid particles (Veyssiere and Khasainov 1994;

Khasainov and Veyssiere 1996). The problem is treated in the frame of the theory of non-ideal detonations, and mass, momentum and heat interphase exchanges are taken into account. Those works have shown that several detonation regimes may exist. These different steady detonation regimes and their structure have been analyzed in detail in Veyssiere and Khasainov (1994), their initiation and stability in Khasainov and Veyssiere (1996). Here, we only sum up the main features of the different regimes. Complete discussion can be found in the abovementioned references. The first detonation regime is the single-front detonation (SFD), which corresponds to a detonation supported by a unique heat release zone involving both gaseous reactions and reactions between particles and gases. In this case, the reaction of particles occurs, at least partially, before the CJ plane so that burning of particles contributes to detonation propagation. On the contrary, when the particles react behind the CJ plane, the detonation is supported only by heat release from gaseous reactions: it is the "pseudo-gas" detonation (PGD). In certain cases, a two discontinuity front structure may exist; the first front is supported by gaseous reactions, the second one by reactions between particles and gases, which is the so-called double-front detonation (DFD). To summarize, the propagation mode was demonstrated to be controlled by the effective heat release rate $dq/dt]_{eff}$ which is the balance between heat release rate (due to both gaseous reactions and reactions between particles and gases) and heat loss rate: this effective heat release rate depends on the size and mass concentration of particles (Veyssiere and Khasainov 1994; Khasainov and Veyssiere 1996). In addition, the possibility of multiple propagation regimes for a given set of initial conditions was established in these studies.

But until now, available data on the influence of suspended particles on the detonability of gaseous mixtures, including detonation initiation, are very limited. As compared with the detonation in the pure gaseous mixture, the coupling between the shock front and the reaction zone is expected to be modified by addition of particles, due to chemical reactions between particles and gases. Therefore, it seems natural to suppose that the cellular structure should encounter changes (in size, regularity, etc). However, the detailed kinetics of reaction of particles with gases is not known and it is impossible, in the absence of experimental data on this subject, to predict whether and how the addition of solid particles may influence the cellular detonation structure. This motivated our study of the conditions of existence and characteristics of the cellular structure in the detonation of hybrid mixtures. Experiments have been done in hydrogen-air mixtures with aluminium particles in suspension, and the dependence of the propagation regime on the reactivity of particles has been investigated. First results displaying the existence of the cellular structure in the case of hybrid mixtures have been reported by Ingignoli et al. (1999b).

2 Experimental conditions

Experiments have been performed in an experimental setup similar to that used previously by Veyssiere (1986). The 69-mm diameter circular cross section detonation tube (see Fig. 2) is disposed vertically and has been lengthened so that the distance available for detonation propagation (between V_1 and V_2 , see Fig. 2) is now about 6 m. Dispersion of particles in the gaseous mixture is achieved by a dust generator using a fluidized bed (Veyssiere 1985). The tube is filled by the flow of the different components from the bottom to the top of the tube. Quasi instantaneous initiation of the detonation is achieved by a blasting cap. Evolution of the characteristic parameters of the detonation wave during its propagation along the tube is recorded by ionization probes, photodetectors and piezo-electric pressure gauges (see Veyssiere 1985, 1986 and Veyssiere et al. 2000). Under these conditions, experimental observations of Veyssiere (1985) had shown that a detonation was formed within a distance less than 1.9 m with a velocity approaching that of the steady detonation by less than 2%, and that a steady detonation wave propagated up to the end of the tube (4.175 m) for the pure gaseous hydrogen air mixtures as well as for the same mixtures laden with aluminium particles. More recent experiments of Veyssiere et al. (2000) in the present 6 m long tube corroborate preceding results and confirm that the build-up process of the detonation initiated in a hybrid mixture by a strong energy source is governed rather by the reactions of gaseous components. This explains why the length to diameter ratio of the tube required to observed steady detonations (here L/d = 86) is smaller than for the case of heterogeneous mixtures where only reactions between particles and gases support the propagation of the detonation front.

The characteristic cellular structure of the detonation regime is recorded on 1-m long metallic soot plates (covering half the circumference of the tube) mounted at the walls in the terminal part of the tube (see Fig. 2).

Three kinds of aluminium particles have been used (see Fig. 3): $3.5 \,\mu\text{m}$ (labelled "A1") or $13 \,\mu\text{m}$ ("A2") atomized particles, and flakes ("F") having a characteristic thickness of $0.5\text{--}1 \,\mu\text{m}$ and different length (up to $45 \,\mu\text{m}$). The latter were supposed to be more reactive than the atomized particles, on account of their large specific area.

Lean, near stoichiometric and rich hydrogen-air mixtures (r = 0.87, 1.06 and 1.32 respectively) have been experimented. Hereafter, the equivalent ratio r is always related to the composition of the pure gaseous mixture. This gaseous equivalent ratio r together with the size and mass concentration of particles is the most pertinent way to differentiate the different mixtures. Indeed, considering direct reaction of aluminium with oxygen, one could define an other equivalent ratio depending on aluminium concentration in the gaseous mixture: in this case, stoechiometry would be achieved for a theoretical aluminium concentration $\sigma = 315 \text{ g/m}^3$. But neither this equivalent ratio relative to aluminium, nor a global equivalent ratio including hydrogen and aluminium are relevant to the problem, since it is worthy to recall that aluminium parti-



Fig. 2. Experimental setup





Mean diameter 13µm

Atomized

Fig. 3. Microphotographies of aluminium particles

cles may react not only with oxygen, but also with water vapor and nitrogen: this means that aluminium particles can burn in the detonation products of a stoichiometric or rich hydrogen-air mixture and may contribute to an additional heat release, whatever the initial equivalent ratio of the gaseous mixture; but this heat addition and its instant of occurrence (which is controlled by the effective heat release rate $dq/dt]_{\text{eff}}$) are strongly determined by the size of particles, as recalled in Sect. 1.3. With the gaseous mixtures used in the present study, the global equivalent ratio is always greater than 1 (even for the lean gaseous



Direction of detonation propagation

Fig. 4. Soot tracks records of the detonation cellular structure in hydrogen-air mixture r = 0.87 without particles

mixture, as soon as the particle concentration is greater than $\sigma = 20 \text{ g/m}^3$) and the size of particles is sufficiently large, so that direct reaction of aluminium with oxygen is unlikely.

3 Results

Firstly, the cellular structure was recorded in hydrogen-air mixtures without particles. Typical record of the structure on a soot plate is shown in Fig. 4, for the propagation of a steady detonation in a mixture having an equivalent ratio r = 0.87. It presents the classical features of the cellular structure in this kind of mixture: the network of cells is fairly irregular with a significant dispersion of the cell dimensions. Particularly, small size cells may be observed inside larger ones. They are located preferably in the first part of the cells of larger size. This kind of substructure has already been observed and described by Manzhalei (1977): it occurs in the detonation of mixtures for which the ratio $E_a/RT_{\rm ZND}$ is larger than 6, where E_a is the activation energy of the global reaction and T_{ZND} the temperature behind the shock front. This is precisely the case of the present gaseous mixture for which, taking account of a value of $T_{\rm ZND}$ equal to 1488 K and a value of E_a equal to 19 kcal/mole as proposed by Miyama and Takeyama (1964), the ratio $E_a/RT_{\rm ZND}$ is of the order of 6.43. This remark being taken into account, the average cell width is determined to be $\lambda = 1.3$ cm. This value is in good agreement with those determined for the same mixture by other works: between the value proposed by Guirao et al. (1982) and that of Cicarelli et al. (1994).

When adding aluminium particles to the same mixture, different changes of the cellular structure can be observed, according to the characteristics of particles. Note that for all experiments the results of which are presented hereafter, the detonation propagated steadily, as explained in Sect. 2, at the place where soot plates are disposed (that



Direction of detonation propagation

Fig. 5. Soot tracks records of the detonation cellular structure in hydrogen-air mixture r = 0.87 with flakes F



Fig. 6. Effect of flakes F on pressure evolution behind the detonation front



Fig. 7. Variation of detonation velocity with a luminium particle concentration for particles A1 and A2, and flakes ${\rm F}$



Direction of detonation propagation

Fig. 8. Soot tracks records of the detonation cellular structure in hydrogen-air mixture r = 0.87 with atomized particles A2



Fig. 9. Effect of atomized particles A2 on pressure evolution behind the detonation front

is, at the upper end of the tube, 5 m after the initiation point, see Fig. 2).

With small particles A1 and flakes F, the cell width becomes smaller than for the pure gaseous mixture and the network is more regular. The example shown in Fig. 5 has been obtained with particles F (similar results have been obtained with particles A1) for a concentration of aluminium particles $\sigma = 220 \,\mathrm{g/m^3}$. Under these conditions, the cell width is $\lambda = 0.80$ cm. Simultaneously, important changes can be observed on the pressure evolution (see Fig. 6): the front pressure is increased and pressure level in burnt products is significantly higher than in the pure gaseous mixture. Analysis of detonation velocity dependence on particle concentration (Fig. 7) indicates that with these two kinds of particles, detonation velocity is increased. According to the preceding results of Veyssiere and Khasainov (1994), this propagation regime is that of a single-front detonation (SFD).

With larger particles A2, opposite behavior is observed. As shown in Fig. 8, the cell size is increased and its regularity becomes poor, with a large dispersion in cell dimensions. Figure 8 has been obtained with a particle concentration $\sigma = 60 \,\mathrm{g/m^3}$. The average cell width for this case is $\lambda = 2.5 \,\mathrm{cm}$. The pressure evolution corresponding to this experiment (see Fig. 9) displays a behavior completely different from that of Fig. 6 : first, the front pressure is hardly changed by addition of particles; then, during the first $100\,\mu s$ in the burnt products, the pressure level remains close to that of the mixture without particles, perhaps slightly less elevated; but beyond this delay, pressure increases again and a second discontinuity front is observed at about 200 µs behind the leading one. At the same time, as shown in Fig. 7, the detonation velocity decreases. This situation has been shown to correspond to a double-front detonation (DFD) (Veyssiere and Khasainov 1994).

Similar observations have been done in the near stoichiometric (r = 1.06) and rich (r = 1.32) mixtures.

4 Discussion

Present experiments in hybrid mixtures (hydrogen-airaluminium particles) demonstrate without ambiguity that the cellular structure exists in this kind of reactive medium. To our knowledge, it is the first time that such an evidence is provided. Obviously, the significant changes observed in the cellular detonation structure in comparison with that of the detonation of the pure gaseous mixture are due to secondary reactions between solid particles and gases. Interpretation of these results should be made in relation with the structure of the different detonation regimes in hybrid mixtures as established by the works of Veyssiere and Khasainov (1994) and Khasainov and Veyssiere (1996). Particularly, it should be kept in mind that, due to the order of magnitude of their characteristic burning time longer than for gases, only part of the heat release due to combustion of particles (possibly none) contributes to the propagation of the leading front, the remaining being responsible of the changes in the flow structure downstream of the detonation front. Thus, the cellular structure remains fundamentally determined by the reactivity of the gaseous components of the mixture. The change of the cell size with the variation of the velocity of the leading front confirms this interpretation: indeed, the average width of the cell structure diminishes, from that of the pure gaseous mixture, when the detonation velocity is increased by heat release addition from particles, and, on the contrary, augments when the detonation velocity is decreased due to heat losses to particles. Regularity of the cellular structure evolves accordingly, following the variation of temperature at the shock front: it becomes more regular when detonation velocity increases, and less regular when the detonation velocity decreases. Beyond, existence of a non-monotonic multistage heat release process with different characteristic kinetic times (which is a fundamental feature of the detonation in hybrid mixture, see Veyssiere and Khasainov 1994) leads to presume the existence of two cellular structures, each of them being related to different kinetic phases of the heat release process. This question will be discussed later in this paper. Let us examine, first, the dependence of the cell size on the velocity of the leading front.

Further analysis of cell size variations can be made by comparing the cell width of the detonation in a hybrid mixture with that in the same gaseous mixture without particles. Let us consider a detonation propagating in a hybrid mixture with a velocity D_p . The corresponding value of the detonation cell width is λ . The detonation in the pure gaseous mixture having the same composition propagates with a velocity D_0 and its cell size is λ_0 . The cell size normalized by the cell size of the pure gas (λ/λ_0) has been plotted in Fig. 10 versus the detonation velocity normalized by the detonation velocity of the pure gas (D_p/D_0) . It can be noticed that the dimensionless value of the cell size decreases monotonically with augmentation of the detonation velocity. The dependence of λ/λ_0 on variations of D_p/D_0 is quite similar to what has been observed by Desbordes (1988) in gaseous mixtures for the dependence of the cell size on the rate of detonation overdrive (see Sect. 1.1). In the experiments of Desbordes (1988), the velocity of the gaseous detonation was changed by generating quasi-steady overdriven detonations. Here, the velocity of the detonation front is augmented or diminished by increasing or decreasing the heat supporting the propagation of the leading front, by means of solid particles. These different manners to vary the velocity of the leading front results in analogous variations of the detonation cell size. Therefore, it appears of interest to use the same form of correlation law to interpret our experimental results. The relationship (2) is proposed:

$$\frac{\lambda}{\lambda_0} = \frac{D_p}{D_0} e^{\frac{E_a}{RT_{\rm ZND}} \left[\left(\frac{D_0}{D_p} \right)^2 - 1 \right]} \,. \tag{2}$$

The value T_{ZND} of the temperature at the leading front in ZND state is that of the pure gaseous mixture. Two different values of the activation energy have been used for the mixture hydrogen-air: $E_a = 19 \text{ kcal/mol}$ (Miyama and Takeyama 1964) and $E_a = 17.2 \text{ kcal/mol}$ (Cheng and Oppenheim 1984). Results of calculations are shown in Fig. 10. As can be seen, we have drawn the values of the relationship (2) for values of $D_p/D_0 > 1$ as well as for values of $D_p/D_0 < 1$. In the case of gaseous mixtures, the validity of (1) had been established only for $D/D_{\rm CJ} > 1$, since only CJ or strong detonation waves can be physically observed. However, there is no reason to limit, a priori, the applicability domain of relationships (1) or (2)to the case of $D_p/D_0 > 1$ as it is only founded on the assumption of proportionality between the cell width and the chemical induction length of gaseous reactions. As indicated by formula (2) this proportionality ratio just depends on the temperature behind the leading shock and on the detonation velocity. Thus, it can be seen in Fig. 10 that for $D_p/D_0 > 1$, there is a good agreement between the correlation curve and our experimental results. When $D_p/D_0 < 1$, two situations are observed according to the value of D_p/D_0 : For $0.98 < D_p/D_0 < 1$, the experimental values fit quite well with the correlation curve, but for smaller values of D_p/D_0 , the predicted values are signif-



Fig. 10. Variation of the detonation cell width λ with the velocity of the leading front in the hybrid hydrogen-air-aluminium particles mixture

icantly larger than the experimental ones (at the upper limit of the accuracy interval of cell width measurement), whatever is the chosen value for the activation energy.

Above results indicate that the correlation law between the cell size and the detonation velocity displayed by Desbordes (1988) should be more universal and valid not only for velocity values larger than that of the self sustained CJ detonation, but also for smaller ones. Further analysis of the detonation structure permits to precise this interpretation. For $D_p/D_0 > 1$, the detonation propagates in the hybrid mixture in SFD regime, that is, the detonation is supported by a unique heat release zone where combustion of gases and particles occurs. Additional heat release, coming from burning of particles, increases the velocity of the detonation front and has the same effect on the detonation cell size as a supported overdriven detonation in the pure gaseous mixture. On the contrary, for $D_p/D_0 < 1$, combustion of particles occurs downstream of the CJ plane, in a reaction zone separated from the gaseous one. Consequently, particles absorb (due to momentum and heat transfer from gas to particles behind the detonation front) part of the heat released in the gaseous reaction zone to heat up to their ignition temperature, which results in decreasing the effective heat release rate (see Sect. 1.3) and the detonation velocity accordingly. In Fig. 10, the case of $0.98 < D_p/D_0 < 1$ corresponds to detonations propagating in hybrid mixtures in the PGD regime. In this case, as demonstrated by Veyssiere and Khasainov (1994), burning of aluminium particles does not contribute to the heat release supporting the detonation propagation, but gives rise to a secondary compression of products in the unsteady flow behind the CJ plane. Thus, one observes the decrease of the detonation velocity and an augmentation of the cell size, in excellent agreement with the correlation law (2). For detonations propagating with a more important velocity deficit $(D_p/D_0 < 0.98 \text{ in Fig. 10})$, the cell dimension predicted by the correlation law (2) is larger than the value measured in experiments. Several reasons may be invoked to seek an explanation for this mediocre agreement. First, contrarily to the case $0.98 < D_p/D_0 < 1$, the detonations for which $D_p/D_0 < 0.98$, correspond to propagation in the DFD regime. However, it remains to investigate in more details the actual influence of the second discontinuity front on the cellular structure. One may also suppose that such velocity deficit could be characteristic of a low-velocity detonation regime (see Veyssiere and Khasainov 1994). In the present state of our knowledge, it is not possible to propose a firm explanation, all the more so because there exists some uncertainty on the actual value of the activation energy of the gaseous reactions.

An other problem arises from the occurrence of secondary heat release due to reactions of aluminium particles with gases. Since the characteristic times of gaseous reactions and reactions between particles and gases differ strongly (possibly by more than an order of magnitude), it is conceivable to assume the existence of a more complicated cellular structure connected to the different kinetic phases of the heat release process: existence of two networks of cells having different characteristic sizes could be conjectured. This assumption has been confirmed recently by the results of Lamoureux et al. (2001) in the detonation of gaseous nitromethane oxygen mixtures, where they have observed two cellular structures of different size, each of them corresponding to a kinetic phase of nitromethane oxydation. However, in the present state of our investigations and with the resolution of our registrations, it was not possible to provide evidence of the existence of a secondary cellular structure in our experimental conditions.

5 Concluding remarks

Our experimental results positively demonstrate that the cellular structure is a "signature" of the detonation, not only in homogeneous gaseous mixtures, but also in hybrid solid particles-gas mixtures. In present experiments, addition of aluminium particles to hydrogen-air mixtures allowed to vary the ratio D_p/D_0 in the range 0.9–1.05. At the same time, the relative cell width λ/λ_0 has been found to vary from 0.5 to 3 times the value of the cell width for the pure gaseous mixture: This should result in a considerable change of the detonability of the mixture. Moreover, the correlation law between the variation of the detonation velocity and the cell size appears to be valid for velocities smaller as well as larger than that of the steady CJ detonation. From this point of view, addition of particles is an interesting way to vary the detonation velocity of a gaseous mixture in the domain surrounding the CJ regime. Depending on particle diameter, it is possible to promote self sustained detonation regimes with detonation velocities either larger or smaller than the CJ detonation velocity of the pure gaseous mixture.

Further investigations under various experimental conditions are needed to precise these first results and to get better understanding of the influence of particles on the cellular structure. Particular attention should be paid to the investigation of the possible existence of two cellular structures of different size or of a substructure (which is relevant to the same problem). Until now, we have not

found on our soot tracks detectable evidence substantiating this point of view. Beyond the experimental difficulties encountered on account of the presence of solid particles, which spoil the soot tracks and weaken the quality of cellular structure registration, a difficulty comes from the ignorance of the dimension of the cellular structure which could result from secondary reactions. The only indications come from the recent results of Ingignoli et al. (1999a) and Zhang et al. (2001) in two-phase mixtures, according to which the the cell dimension would be of the order of magnitude of a few tens of centimeters. An other possibility is to get an estimation of the cellular structure dimension from numerical simulations. This is very important to predict the pertinent dimensions of the experimental configuration necessary to perform relevant experiments. Two-dimensional numerical simulations of the structure of hybrid detonations in hydrogen-airaluminium particles are under development for this purpose.

Acknowledgements. The present work has been done with the support of INTAS under grant no. 97-2027

References

- Borisov AA, Khasainov BA, Veyssiere B, Saneev EL, Fomin IB, Khomik SV (1991) On the detonation of aluminiumair and oxygen suspensions In: Borissov AA (ed) Dynamic structure of detonations in gaseous and dispersed media. Kluwer Academic Publishers, pp 215–253
- Cheng RK, Oppenheim AK (1984) Autoignition in methanehydrogen mixtures. Combustion and Flame 58, 125–139
- Cicarelli G, Ginsberg T, Boccio J, Economos C, Sato K, Kinoshita M (1994) Detonation cell size measurements and predictions in hydrogen-air-steam mixtures at elevated temperatures. Combustion and Flame 99(2), 212–220
- Denisov YN, Troshin YK (1960) Structure of gaseous detonations in tubes. Z Tekh Phys 30, 450–459
- Desbordes D (1988) Transmission of overdriven plane detonations: critical diameter as a function of cell regularity and size. AIAA Progress in Astronautics and Aeronautics, Ed. AIAA New-York, vol 114, pp 170–186
- Gardner BR, Winter RJ, Moore MJ (1986) Explosion development and deflagration-to-detonation transition in coal dust/air suspensions. 21st Symposium (International) on Combustion, the Combustion Institute, pp 335–343
- Guirao C, Knystautas R, Lee JHS, Benedick W, Berman M (1982) Hydrogen-air detonations. 19th Symposium (International) on Combustion. The Combustion Institute, Pittsburgh, pp 583–590
- Ingignoli W, Veyssiere B, Khasainov BA (1999a) Study of detonation initiation in unconfined aluminium dust clouds. In: Gaseous and Heterogeneous Detonations, ENAS Publishers, Moscow, ISBN 5-89055-016-0, pp 337–350
- Ingignoli W, Veyssiere B, Khasainov BA (1999b) Detonation of Hydrogen-Based Gaseous Mixtures With Suspended Aluminium Particles. Proceedings of the 17th International Colloquium on Gasdynamics of Explosions and Reactive Systems, Heidelberg, July 25–30, CD-Rom ISBN 3-932217-01-2, paper no 151

- Kauffman CW, Wolanski P, Arisoy A, Adams PR, Maker BN, Nicholls JA (1984) Dust, hybrid, and dusty detonations. Progress in Astronautics and Aeronautics, Ed. AIAA New-York, vol 94, pp 221–240
- Khasainov BA, Veyssiere B (1996) Initiation of detonation regimes in hybrid two-phase mixtures. Shock Waves 6, 9–15
- Lamoureux N, Matignon C, Sturtzer M-O, Desbordes D, Presles HN (2001) Interprétation de la double structure observée dans l'onde de détonation du nitrométhane gazeux. Comptes rendus Acad Sci Paris t 329, Série IIb, pp 687–692
- Li YC, Alexander CG, Wolanski P, Kauffman CW, Sichel M. (1993) Experimental investigations of accelerating flames and transition to detonation in layered grain dust. AIAA Progress in Astronautics and Aeronautics, Ed. AIAA New-York, vol 154, pp 170–184
- Manzhalei VI (1977) Fizika Goreniya i Vzryva 3, 470-473
- Miyama H, Takeyama T (1964) J Chem Phys 41, 2287
- Peraldi O, Veyssiere B (1986) Experimental study of the detonations in starch particles suspensions with O₂/N₂, H₂/O₂, and C₂H₄/O₂ mixtures. AIAA Progress in Astronautics and Aeronautics, Ed. AIAA New-York, vol 106, pp 490–504
- Pu YK, Li KY, Jarosinski J (1997) Transition to detonation in aluminum dust-air mixtures under weak ignition conditions. Proceedings of the 16th ICDERS, Wydawnicto "akapit" ed, Krakow, Poland, pp 259–262
- Strauss WA (1968) Investigation of the Detonation of Aluminium Powder-Oxygen Mixtures, AIAA J 6(9), 1753–1756
- Tulis AJ, Selman JR (1982) Detonation tube studies of aluminum particles dispersed in air. 19th Symposium (International) on Combustion, the Combustion Institute, pp 655-663

- Veyssiere B (1985) Double-front detonations in gas-solid particles mixtures. AIAA Progress in Astronautics and Aeronautics, Ed. AIAA New-York, vol 94, pp 264–276
- Veyssiere B (1986) Structure of the detonations in gaseous mixtures containing aluminium particles in suspension. AIAA Progress in Astronautics and Aeronautics, Ed. AIAA New-York, vol 106, pp 522–544
- Veyssiere B, Khasainov BA (1994) Structure and multiplicity of detonation regimes in heterogeneous hybrid mixtures. Shock Waves 4(3), 171–186
- Veyssiere B, Ingignoli W, Khasainov BA(2000) On the possibility to modify the performance of propellant mixtures used in RAMAC by addition of metallic particles. Journal Phys IV 10, 99–107
- Wolanski P (1991) Deflagration and detonation combustion of dust mixtures. Progress in Astronautics and Aeronautics, Dynamics of deflagrations and reactive systems: heterogeneous combustion, Ed. AIAA New-York, vol 132, pp 3–31
- Zhang F, Grönig H (1991) Transition and structure of dust detonations. In: Borissov AA (ed) Dynamic structure of detonation in gaseous and dispersed media, Kluwer Academic Publishers, pp 157–213
- Zhang F, Grönig H (1993) Detonability of organic dust-air mixtures. Progress in Astronautics and Aeronautics, Dynamic aspect of explosion phenomena, Ed. AIAA New-York, vol 154, pp 195–214
- Zhang F, Greilich P, Grönig H (1992) Propagation mechanism of dust detonations. Shock Waves 2 81–88
- Zhang F, Grönig H, Van De Ven A (2001) DDT and detonation waves in dust-air mixtures. Shock Waves 11, 53–71