Investigation of Detonation Initiation in Aluminium Suspensions

B. Veyssiere, B.A. Khasainov, A. Briand

Laboratoire de Combustion et de Détonique, UPR 9028 CNRS,
ENSMA, BP 40109, 86961 Futuroscope, France

1 Introduction

The possibility for suspensions of metallic solid particles dispersed in an oxidizing atmosphere to detonate has been considered for a long time, on account of explosion hazards in industry, but also for propulsion and military applications. However detonation initiation in solid particle-gas two phase mixtures is well known to be a difficult problem due to the weak detonability of such mixtures in comparison with homogeneous gaseous mixtures. Strong initiation by a shock wave is required to minimize the distance of transition to self-sustained steady detonation. Here, we investigate numerically the critical conditions for detonation initiation in aluminium/ oxygen and aluminium/air mixtures in connection with the detonation cellular structure in two-phase solid particle gas mixtures.

2 Initiation of plane detonations

Calculations have been carried out with the numerical code already used to investigate detonation initiation and propagation in homogeneous and heterogeneous two-phase reactive mixtures. Details of numerical modelling have been described in [1]. Point source explosion is used to initiate the detonation. Different kinds of particles uniformly dispersed in oxygen have been considered: atomized particles, the shape of which was assumed to be spherical, with diameters 2 µm, 3.5 µm, 5 µm, 8.6 µm and 13 µm respectively; and “flakes” particles which are supposed to have a burning rate higher than that of atomized particles. It is worthy to note that the mass of flakes is equivalent to that of spherical 8.6 µm particles. Particle mass concentration σ was varied between 0 and 4000 g/m^3 (stoichiometric concentration of aluminium in oxygen is about 1500 g/m^3).

In the case of a tube with infinite diameter, which corresponds to an ideal CJ plane detonation, we have verified that the detonation parameters calculated with our model, when steady propagation has been reached, coincide with the thermodynamical CJ values. However, particle size influences the length of the reaction zone.

In the case of finite tube diameter, friction and heat transfer to the tube walls in the reaction zone are taken into account, according to phenomenologic laws as described in [1]. Calculations have been made for different tube diameters and particle sizes.

When comparing (see Fig.1) the experimental values of detonation velocity registered by Strauss [2] with our calculations in corresponding conditions (tube of 26.4 mm internal diameter; atomized 5µm particles and flakes), one can conclude that the observed velocity deficit is qualitatively explained satisfactorily by the heat losses to the tube walls. However, calculated values are higher than experimental ones for corresponding values of particle diameter, so that values reported by Strauss fit better with larger particles (13µm). This can probably be explained by the fact that during experiments of Strauss the glass tube used to contain the reactive mixture is burst just after the passage of the detonation wave, within a characteristic distance which is smaller than the

Correspondence to: veyssiere@lcd.ensma.fr

reaction zone thickness. For that reason, rapid expansion of products behind the detonation front could induce an augmentation of energy loss rate and thus a decrease of detonation velocity.

3 Initiation of spherical detonations

Numerical simulations have been performed in 1D spherical configuration of shock to detonation transition, for a propagation radius of the leading shock front ranging between 0 and 4 meters. The characteristics of particles and their concentration in oxygen were the same as for the plane case. An example of dependence of the critical initiation energy on the concentration of particle is displayed in Fig.2 for 5μm atomized particles. According to these results, the critical initiation energy for 5μm particles at stoichiometric concentration (1500 g/m^3) should be $2.5\cdot10^4$ J, which is about 5.5g of TNT.

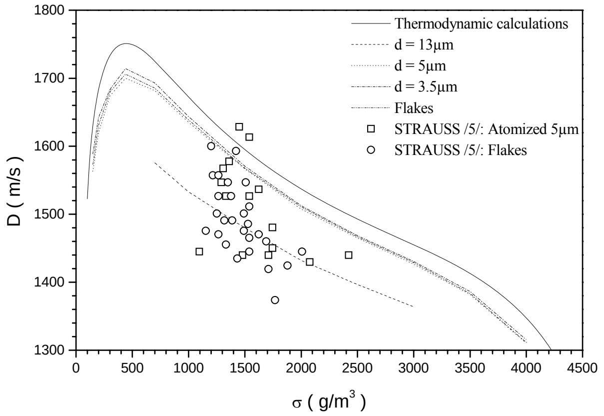

Fig.1 – Comparison of calculated values of detonation velocity in aluminium/oxygen mixtures contained in 26.4mm diameter tube with the experimental values measured by Strauss [2]

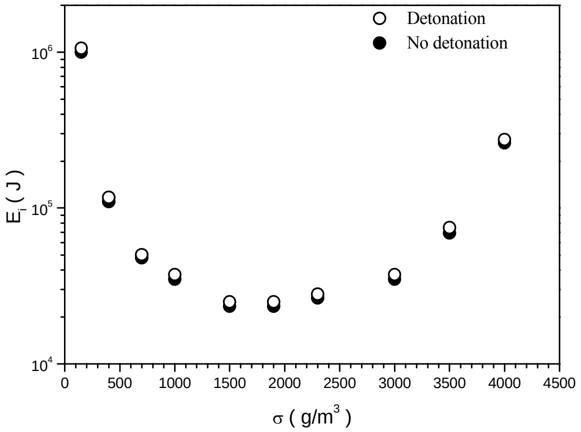

Fig. 2 – Calculated critical energy for initiation of spherical detonation in aluminium/oxygen mixtures for $d = 5$ μm particles

Because of the apparent similarity with detonation initiation in gaseous mixtures, it is worth examining whether semi-empirical criteria derived for gaseous mixtures can be transposed to heterogeneous gas-particle mixtures. Assuming an ideal point explosion source, the critical radius for detonation initiation R_c, according to the results of Benedick et al. [3], should be equal to the characteristic explosion length R_0. Desbordes [4] displayed that in the case of critical initiation of spherical detonations, the critical predetonation radius R_c could be correlated to the detonation cell size λ, according to the relationship $R_c = 20\ \lambda$. This leads to propose an estimate of the critical initiation energy as: $E_c = 8000\ p_0\cdot\lambda^3$.

The only available experimental data on the cellular structure in aluminium-oxygen rich mixtures are those reported by Ingignoli et al. [5]. They have recorded in aluminium flakes suspended in oxygen mixtures a few cellular-like structures with a characteristic dimension of 5-10 cm. It follows that the critical predetonation radius would be of the order of 1 m and the critical initiation energy $E_c = 10^5$ J. This value of E_c lies between that derived from numerical simulations ($2.5\cdot10^4$J), and the initiation energy value used by Ingignoli et al. [5] in the experiments in which they observed the cellular structure ($6.78\cdot10^5$ J). As compared with the critical energy proposed by Matsui [6] for hydrocarbon-oxygen mixtures (10^{-3} to 1 J) and by Makeev et al. [7] for stoichiometric hydrogen-air ($4{\cdot}10^3$ J), present results confirm the weak detonability of two-phase solid particle-gas mixtures.

In aluminium-air mixtures, Zhang et al. [8] have displayed in their experiments that the detonation cell size is of the order of magnitude of 40 cm. When applying present analysis to aluminium - air mixtures, the corresponding values of the predetonation radius and critical initiation energy come out to R_c = 8m and $E_c = 5{\cdot}12\ 10^7$ J, that is more than 11 kg of TNT. This would explain why Tulis [9] was unsuccessful to observe detonation formation in his attempts to detonate unconfined aluminium-air dust clouds: his initiating charge was only 2.27 kg of solid explosive and the radius of his cylindrical cloud was 3m. Even in his elongated 10-m long clouds it was unlikely to initiate a detonation because of the insufficient lateral size (1 m) of the cloud which caused early lateral expansion of the combustion products, preventing the coupling between the leading shock front and the reaction zone, necessary for the detonation build-up.

4 Numerical modelling of the detonation cellular structure

Above analysis indicates the need to take account of the multi-dimensional character of the detonation wave to get a more precise estimation of the critical conditions for detonation initiation in gas-solid particle mixtures. This requires a better knowledge of the cellular detonation structure in gas-solid particle two-phase mixtures, and particularly of its characteristic size. However, the available experimental data are very scarce: as mentioned above for aluminium - air or - oxygen mixtures only very limited observations have been reported in [5] and [8]. To get relevant results on detonation initiation in these two-phase mixtures, it is necessary to perform experiments in volumes of large dimensions (see section 3). On account of the difficulty to perform such large scale experiments, we have investigated this problem numerically.

Numerical simulations have been made with the computer code developed on the basis of the model described in [1]. To get reliable description of the cellular structure, one key problem is to incorporate an as realistic as possible model of aluminium combustion kinetics. Similarly to the case of gaseous mixtures, the reaction zone can be divided in two parts: an induction period and a reaction period. The induction period has been modelled by an Arrhenius law with E_a = 17000 cal/mol, according to the results of Merzhanov [10] on aluminium wires ignition in oxygen. The reaction period corresponds to the burning of particles and is monitored by their combustion time. A classical d^2-regression law is used for the combustion time: $\tau_b = kd_0^2$ (with d_0 particle initial diameter, k burning time constant). But these characteristic times are completely different from those in gaseous homogeneous mixtures: particularly, depending on particle size the reaction time can become larger than the induction time.

Initial conditions of numerical simulations have been chosen so that to match the existing experimental results. Since in the experiments flakes had been used, an effective diameter of particles has been estimated to perform numerical simulations: it corresponds to the same mass for an individual particle. But the drag coefficient has been changed to take account of the particle shape, and the burning constant is set to $k = 1{,}6{\cdot}10^6$ s/m², according to Ingignoli results [11], to match the higher reactivity of flakes. For aluminium-air mixtures [8] we have considered a particle mass concentration σ = 500 g/m^3 (richness Φ = 1.61) and an equivalent particle diameter of 13.5 μm, while for oxygen-air mixtures [5] σ = 1500 g/m^3 and the equivalent particle diameter is 8.6 μm.

Example of 2D-simulation results is displayed in Fig.3 for the case of the aluminium-air mixture. The characteristic cell width is 37.5cm, which is very close to the average dimension of experimental cells reported by Zhang et al. [8]. It is important to underline that this calculated cellular structure has been obtained for a tube of 1.5 m diameter after detonation propagation over 20 m when detonation became self-sustained and its cell size independent on run distance.

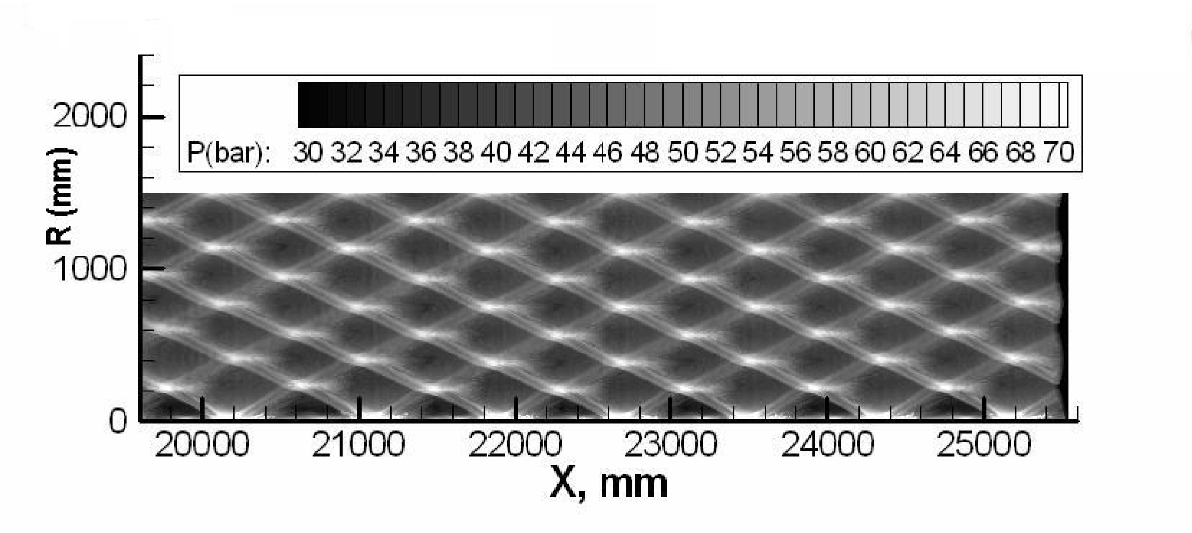

Fig. 3 – Calculated cellular detonation structure in a rich aluminium - air mixture. particle diameter d = 13.5 μm, mass concentration σ =500 g/m^3

Similar calculations have been performed for aluminium-oxygen mixtures. Preliminary simulations with a given set of the kinetic parameters for the induction and reaction periods lead to the value λ=7.5cm for the cell width, that is of the same order of magnitude as λ measured by Ingignoli et al. [5]. Before concluding about the validity of the kinetic model, further investigations should be made to precise the respective influence of the induction and the reaction period on the value of the cell size. It is important to underline that, like for aluminium – air mixtures, this cellular structure has been obtained after a long propagation of the detonation front (over 10 m in a tube with 0.2 m radius). Numerical simulations of the cellular structure in aluminium-oxygen mixtures have also been performed by Benkiewicz and Hayashi [12] and by Khmel and Fedorov [13], but the detonation run distance was much shorter so that it is doubtful that the steady self sustained propagation regime had been reached in those calculations.

Details of the detonation wave structure in the aluminium-air mixture can be drawn from the examination of Fig.4. Fig. 4-a shows pressure P/P_{cj} and gas temperature T/T_{cj} profiles (respectively in the upper part and the lower part of the picture) in a 0.40 m tube at t = 9.34 ms. Fig. 4-b shows the corresponding particle concentration σ/σ_0 and particle temperature $T_s/500$ profiles (respectively in the upper part and the lower part of the picture) at the same instant. Fig. 4-c and 4-d show the same profiles at a subsequent instant t = 9.52 ms. Here, in the case of aluminium particle - air detonations, the lowest temperatures areas behind the detonation front are located in high pressure zones. In contrast, high temperature zones are located just behind the low pressure areas of the detonation front. This indicates that chemical reactions do not occur in high pressure zones as in homogeneous gaseous detonations. An explanation can be proposed from examination of particle concentration and temperature profiles: in high pressure zones around the triple points, the mass particle concentration is the highest; thus it is likely that in such zones, particle ignition temperature is more difficult to reach than in low pressure zones. Indeed, heating of particles up to the critical conditions for their ignition is governed by the balance of heat release and heat losses between gases and particles. Due to the very high concentration of particles in the vicinity of the triple points, particles can ignite within a shorter delay in low pressure zones where the particle concentration is smaller. This feature, which is specific to heterogeneous solid particle mixtures, may be attributed to the role of transverse waves in convecting solid particles toward high pressure zones.

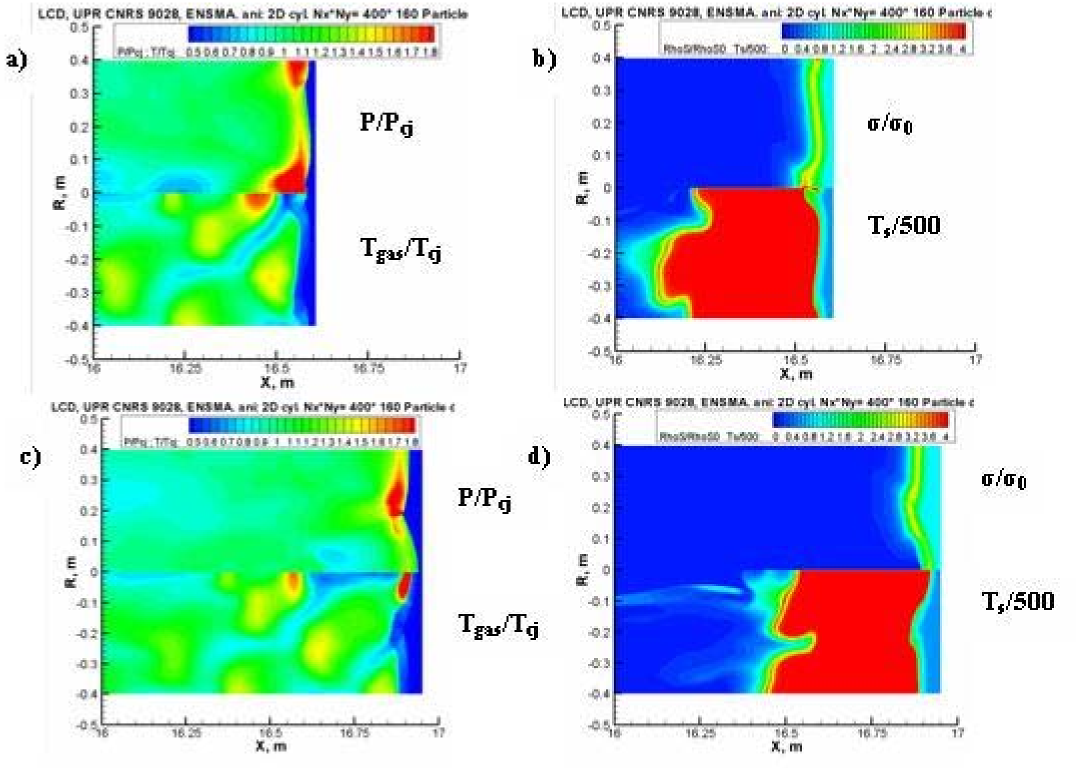

Fig. 4 – Pressure P/P_{cj}, gas temperature T/T_{cj}, particle temperature $T_s/500$ and particle concentration σ/σ_0 profiles for rich Al / air mixture, at two different instants of detonation propagation: t = 9.34 ms and t = 9.52 ms. P_{cj} = 24.5 bar; T_{cj} = 3639 K

5 Concluding remarks

Numerical simulations provide values of the detonation cell size which are consistent with the existing experimental values for aluminium – air and aluminium – oxygen mixtures. This agreement substantiates our estimations of the energy required to initiate a detonation in these mixtures. However, new calculations should be performed with a higher numerical resolution. Further investigations should be done to test different models of aluminium particle ignition and combustion, particularly the influence of the burning constant in the d^2 burning law. Thus, it would be possible to predict more reliably the proper dimensions required to observe self sustained detonations in aluminium –air or – oxygen mixtures.

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