SHOCK-INITIATION AND DETONATION EXTINCTION IN HOMOGENEOUS OR HETEROGENEOUS EXPLOSIVES: SOME EXPERIMENTS AND MODELS

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1 Introduction

Chemical explosives can be found in the gaseous, liquid or solid phases but it is customary to distinguish homogeneous explosives from heterogeneous ones. Indeed experiments have revealed that the dynamical behaviors of detonative or quasi-detonative decomposition regimes of explosives have more to do with the latter classification than with the former. However, distinguishing between homogeneous and heterogeneous behaviors can be questionable depending on the nature, the amplitude or the range of the solicitation applied to the considered explosive.

The objective of the presentation is to report on some recent modeling of critical dynamical behaviors of homogeneous or heterogenous explosives in conditions of chemical decomposition close to the detonation regime. The approaches are based on continuum and micromechanical models. The first part of the presentation will be devoted to the shock-to-detonation transition (SDT) in homogeneous materials, the second one to detonation critical diameters of heterogeneous materials. Each part will be preceded by a short review of experimental facts. The choice is to relate modeling and experiments by means of as simple as possible assumptions, yet mathematically and physically consistent with the underlying dynamics of the phenomena.

2 SDT process in homogeneous materials

The shock-to-detonation process in homogeneous materials has been identified in such impact experiments as those of Campbell et al. (1961a) or Leal-Crouzet (1998) in the liquid explosive nitromethane. It appears to be identical to one of the two observed in the shock-tube experiments by Meyer and Oppenheim (1971) in gaseous mixtures of hydrogen and oxygen : At first, a shock-induced thermal explosion occurs in the close vicinity of the impactor, then a detonation builds up very rapidly in the shock-compressed material (the so-called superdetonation), and, finally, after the superdetonation catches up with the shock, the relaxation towards the normal self-sustained detonation regime is realized. Small deviations from this scenario have been reported and can be attributed to such defects as piston roughness or presence of bubbles in the liquid. A different picture, that essentially features a more gradual detonation build-up, i.e., without bulk thermal explosion and superdetonation, is typical of most solid heterogeneous explosives at moderate shock-pressure solicitations (e.g., Campbell et al. 1961b) but the heterogeneous material will react according to the homogeneous scenario or so at higher shock pressures (e.g., Bowden and Yoffe 1952, Dremin et al. 1970).

A model for the explosion stage of the SDT process in homogeneous explosives will be presented, based on reactive gas-dynamics equations for inviscid fluids (Vidal and Khasainov 1999a-b). The approach was devised so as to be simply applicable to

both gaseous and condensed explosives, and to extend the predictive ability of existing asymptotics-based works to a broader variety of solicitations than previously considered, e.g., noncompressible or compressible, planar or curved, constant- or variable- speed pistons. Closed-form expressions for induction times and critical constraints for shock initiation of adiabatic explosion for arbitrary (i.e., unspecified) shock dynamics are obtained. Here, the shock dynamics appears to be described by the shock normal velocity, normal acceleration and total curvature. The approach amounts to formulating the induction stage of the chemical decomposition process as a Cauchy (initial-value) problem, with the shock taken as the data-surface, and to expanding the solution along material path until the time of explosion. Its key ingredients are (i) a local shock-expansion method, for relating the induction time to the shock dynamics, (ii) the solution to the Cauchy problem for the shock-transformation, for relating the shock dynamics to the rear boundary conditions (the so-called "shock-change equations", e.g., Fickett and Davis 1979), and (iii) analyses of specific rear-boundary conditions, for determining the piston dynamics, then the shock dynamics, in terms of the user's parameters. It is important to acknowledge that this approach is really a synthesis of known ideas, which, however, have not yet been sufficiently emphasized. In particular, the failure criterion is, in some sense, akin to the so-called "breakdown" phenomena postulated by Dremin (1970). Similarly, the solution to the Cauchy problem for the shock transformation, which is known in its elementary form for more than one century, was already used by Jouguet (e.g., Jouguet 1917), for studying detonation-front acceleration, and by Cowperthwaite and Tarver (1975), for interpreting experiments of impact by constant-speed piston. The calculation procedure of the solution to the Cauchy problem is a classical exercise of the theory of (systems of) hyperbolic equations.

A first application is the impact by an incompressible piston. The shock initial dynamics is trivially determined by that of the piston. The predictions of the model are found to be in good agreement with those obtained by means of asymptotics techniques for the case of a constant-speed piston in ideal gases (e.g., Clarke and Kant 1985, Jackson and Kapila 1985, Blythe and Crighton 1989), as well as with our direct numerical simulations for the cases of constant or variable-speed pistons, including the critical conditions.

Another application is the cylindrical or spherical shock-tube problem with an inert high-pressure chamber (HPC) and a reactive low-pressure chamber (LPC). The HPC/LPC interface constitutes the piston that drives the shock induced in the reactive LPC by the expansion of the inert HPC. The calculation of the initial dynamics of the interface constitutes the main difficulty of the problem, due to the existence of an expansion wave departing from this interface inside the HPC. Finally, the result is the critical (minimum) size of a nonideal energy source below which the shocked LPC will not explode (Vidal et al. 1999c).

3 Detonation critical diameter in heterogeneous materials

The modeling of detonation processes in heterogenous explosives faces with four problems.

The first one is to select the essential physical constraints among the many a-priori involved in the flow. Examples are drags, heat transfers and friction phenomena between grains and binder or at crystal cracks, collapse mechanisms, ...). Obviously, these are very dependent on the considered explosive material : aerodynamical drags of particles are an essential property of particle-laden gaseous mixtures but, clearly, are not relevant to bubbly liquids.

The second one is to synthesize the rather large amount of experimental data. Its solution provides a sensible target for modeling and, as a consequence, for assessing the validity of the choice of the retained physical constraints and of their mathematical description.

The third one is the poor knowledge of the constitutive relationships and phenomenological constants describing the state properties of the explosive components, under the high dynamical pressure and temperature conditions that are specific to detonating solid heterogeneous explosives. So far the solution has been the use of crude assumptions on state dependencies of thermodynamical coefficients or, on the contrary, the use of sophisticated equations of state, which involves many adjustable, but physically vague, coefficients.

The fourth one is to decide whether the microscopic fluid properties are sufficiently well described by an isolated cell, representative of the heterogeneity and of its interaction with the surrounding material, or if cooperative phenomena among different cells should also be accounted for.

Other problems, such as sensitivities to initial conditions (e.g., the temperature) are often disregarded.

The variety of behaviors observed in experiments and the mathematical difficulties encountered in solving the many differential equations describing heterogeneous flows have left theoreticians with few hope to obtain sufficiently simple, yet representative, results or trends that could be used as guidelines in physical or numerical experiments. So-called direct numerical simulations have thus been privileged for studying detonation processes in heterogeneous media. This choice however implies solving typically over a hundred coupled differential equations; it does not necessarily allows one to extract physical trends and often ends up in a sophisticated fitting exercise of a small set of experimental results. In addition, sensitivity studies of the modeling on the phenomenological

constants are seldom conducted.

Yet, some progress can be made, based on preliminary synthesis of literature. Thus, as an example of answer to problem #2, an analysis of a broad amount of experimental results on run distance to detonation (RDD) and detonation critical diameter (DCD) will be summarized (Presles et al. 1995, Khasainov et al. 1997). The synthesis extracts a correlation between the RDD, or the reciprocal of DCD, and the reciprocal of the initial *specific surface area* of heterogeneities (i.e., the total hot-spot surface per unit of volume), valid for most liquid heterogeneous explosives. This confirms the idea of Price (1970) that DCD can be a good indicator of the shock sensitivity of explosives. Then, the presentation will focus on an experimental and numerical study on two-dimensional steady detonation properties in a model heterogeneous explosive made of liquid nitromethane (NM) and of glass microballoons (GMB), such as DCD, shock curvature-detonation velocity and detonation velocity-charge diameter relationships (the "diameter effect"). The advantage of such compositions is that their microstructure can be varied in a wide range, for example by changing the GMB size and concentration, and more easily controlled than that of classical porous explosives. Also their modeling is, in some aspects, facilitated because only one physical phenomenum is be accounted for, specifically the viscoplastic collapse of the GMB and the subsequent conductive heat transfer to the adjacent layer of NM.

Thus, a viscoplastic pore collapse model of spherically imploding GMBs (e.g., Khasainov et al. 1993) was implemented in a three-phase formulation of the balance laws (shocked unreacted NM, NM detonation products and GMBs) for mass, momentum and energy for reactive flows under the assumption of a quasi-onedimensional flow. These balance laws were then integrated between the detonation shock-front and the sonic locus (the rear boundary for self-sustained detonation) to obtain the shock normal velocity (D_n) as an eigenvalue for a given total curvature (C) of the detonation shock front (Bdzil 1981, Bdzil and Scott-Stewart 1988). The D_n-C relationship was then used to describe the diameter effect. A semi-quantitative agreement of the model with the experiments is obtained (Ermolaev et al. 1995, Bouton 1997, Bouton et al. 1999). In particular, one important result is that the experimental linear correlation between DCD and reciprocal of the GMB initial specific surface area are modeled, as well as experimental properties of bimodal compositions. Another important result is that the so-called "shock sensitivity reversal effect" (SSRE) (Moulard 1985, 1989) is captured. This effect is characterized by a nonmonotonic dependency of RDD and DCD on the grain size, given the mass fraction of heterogeneities in the composition (i.e., given the specific surface). More specifically, at larger shock pressure, smaller grains are more efficient hot-spots than larger ones, and the larger the shock pressure the larger the number of efficient hot-spots, regardless their diameters. On the contrary, at smaller shock pressures, larger grains are more efficient than smaller grains, and at too small shock pressure only larger grains will be efficient. In the case of our mixtures, we indeed observed in our calculations that the induction time of NM around collapsing smaller GMBs can be too large, that is comparable to the travel time of the fluid particle from the shock to the sonic locus, when considering lower shock pressures. Thus only larger GMBs remains efficient in transferring heat to the shocked unreacted NM due to the relative increase of the conductive heat-dissipation rate in the GMBs compared to the viscoplastic heat-production rate around the GMBs. For monomodal compositions, the calculated SSRE is characterized by a reasonable fit of the experimental linear correlation between DCD and reciprocal of GMB initial specific area only when larger-diameter GMBs are used. As a consequence, the calculated critical diameter appears as a nonmonotonic function of the GMB initial initial diameter, given the specific area, which exhibits a minimum at intermediate values of GMB initial diameter. For bimodal compositions, the calculated SSRE is characterized by a D_n-C curve that exhibit two critical points. The part of the curve below the upper critical point is close to the D_n-C curve of the monomodal composition made of the larger GMBs. Thus, in this case of bimodal distribution of size of potential hot spots, smaller hot spots do not contribute to the chemical-reaction growth at large detonation velocity deficits (i.e., at low shock pressures) so that, again, the critical detonation diameter nonmonotonically depends on the initial specific surface area of the heterogeneous composition.

It is interesting to observe that the increase in efficiency of all hot-spots at high shock-pressure, due to decrease in induction time around the grains, is consistent with the observation that heterogeneous compositions can behave as homogeneous ones for such solicitations (cf. § 1 and 2). It also indicates the existence of a critical hot-spot size, similar, in some sense, to that obtained in the second application described in §2. These results promote the idea that simplifications and syntheses are worthwhile preliminary steps before attempting more formal and expensive treatments of heterogenous fluid flows : on one hand the validity of assumptions on physical constraints and equations of state can be assessed in a relatively easy way, and, on the other, physical trends and interpretations can be obtained.

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