ON THE SHOCK SENSITIVITY REVERSAL EFFECT : THE CASE OF NITROMETHANE SENSITIZED BY MONO- AND BIMODAL GLASS MICROBALLOONS

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Introduction

In order to study the effect of microstructure of heterogeneous explosives on their shock sensitivity, we measured critical detonation diameters and shock front curvatures in nitromethane (NM) sensitized by mono- and bimodal thin-walled glass micro-balloons (GMB). The advantage of such heterogeneous mixtures is that their porosity, void size distribution and specific surface area can be varied in a wide range by changing the GMB size and concentration, and more precisely and easily controlled than that of porous solid explosives.

Experimental results were compared with numerical ones obtained under the assumption that hot spot initiation is controlled by viscoplastic GMB collapse. This mechanism was coupled with the surface burn concept for reaction growth and was implemented in the steady curved critical detonation model for condensed high explosives (HE) (e.g., Yao and Stewart [1]). A satisfactory semi-quantitative agreement of the experimental and numerical results is obtained. The model also explains why our bimodal GMBs did not result in a nonmonotonous experimental dependence of critical diameter on the mean heterogeneity size similar to that observed by Moulard [2] in PBX with bimodal HE grains, and qualitatively interpreted by Khasainov et al. [3]. Indeed, the model predicts the reversal of shock sensitivity (here characterized by the critical diameter, e.g., Price [4]) in bimodal mixtures when the finer GMBs are significantly smaller (about 5 μm) than in the experiments. Another interesting prediction is that the shock sensitivity reversal effect would also be observed with monomodal mixtures if the GMB size is decreased from 50-100 μm to a few μm (Bouton [5]).

Experimental results

The mass fraction of mono- or bimodal GMBs in the mixtures was 1% (in the bimodal case, fine and coarse GMB fractions were equal 0.5%). The GMB diameters were 47 and 102 μm. The mixtures were confined in steel or polyvinylchloride (PVC) tubes. Detonation velocities were measured by means of ionization gauges, and the shock shapes were obtained from streak camera records of the detonation front interaction with an enlightened mirror.

Figure 1 summarizes the dependence of the steady detonation velocity (D_T) on the reciprocal of the charge diameter (d) for mixtures of NM with mono- and bimodal GMBs confined in steel tubes. Figure 2 shows similar results for PVC tubes. All dependencies have the downward concavity that characterizes most solid heterogeneous explosives. The concavity is stronger when the mixtures are confined in PVC tubes. Given a tube diameter, the detonation velocity is higher for monomodal mixtures with smaller GMBs. The GMB volume fraction (Φ_1) is a decreasing function of the GMB diameter (for a constant GMB mass fraction) so that the mass of NM is smaller in mixtures made with larger GMBs. The $D_T(d^{-1})$ curves for steel tubes are quasi-linear at large diameters so that a good estimate of the planar detonation velocity D_∞ can be obtained by extrapolating $D_T(d^{-1})$ to infinite diameter. For a given velocity, it is found that the detonation front in mixtures with larger GMBs has a smaller curvature and supports larger velocity deficits and that, within the experimental accuracy, the nature of the confinement does not practically affect the axial curvature-velocity relationships.

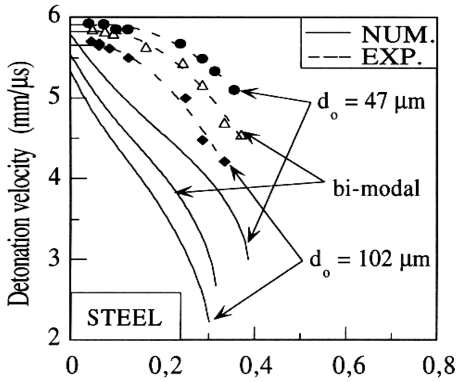

FIG.1.- Reciprocal of charge diameter (mm^{-1})

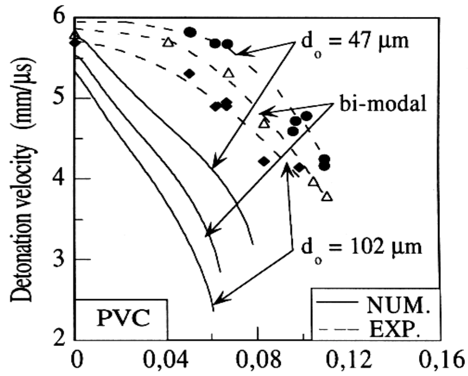

FIG.2.- Reciprocal of charge diameter (mm^{-1})

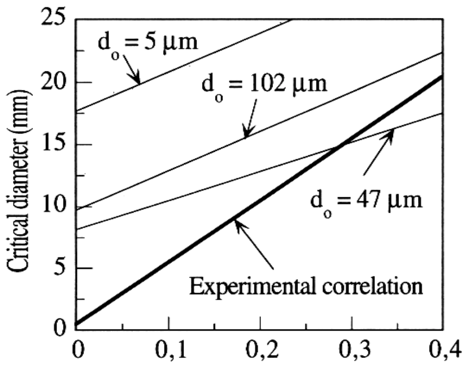

FIG.3.- Reciprocal of specific surface (mm)

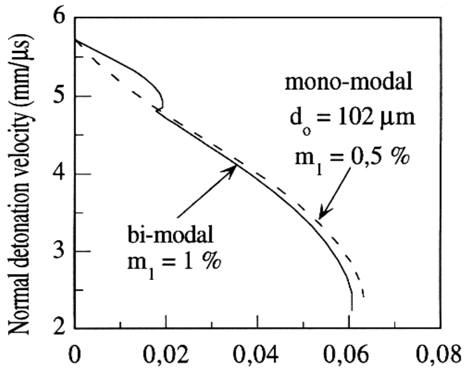

FIG.4. - Shock total curvature (mm^{-1})

The Table below lists the main results which reveal that the smaller the confinement density the higher the critical diameter, that the smaller the initial density ρ_0 of the mixture, the smaller the planar detonation velocity and that the properties of the studied bimodal mixtures are always comprised between the corresponding properties of the monomodal mixtures.

	Monomodal (47 μm)	Monomodal (102 μm)	Bimodal (47 & 102 μm)
ρ_0, (g/cm^3)	1.09	1.04	1.064
Φ_1	0.042	0.085	0.064
D_∞, (mm/μs)	5.914	5.686	5.829
d_{crit} (mm) in PVC, exp./num.	8.5/10.8	9.2/13.9	8.5/13.3
d_{crit} (mm) in steel, exp./num.	3.0/2.5	3.0/3.2	3.0/2.7

Numerical results and discussion

The viscoplastic pore collapse model of spherically imploding GMBs 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 shock and the sonic locus to obtain the shock normal velocity (D_n) as an eigen-value for a given total curvature (C) of the shock front of self-sustained detonation. The D_n-C relationship was then used to describe the diameter effect curves (Bouton [5], Ermolaev et al. [6]).

The numerical and experimental critical diameters d_{crit} for steel and PVC tubes are given in the Table. Figures 1 and 2 compare the numerical and experimental D_n-$1/d$ relationships respectively for steel and PVC tubes. A semi-quantitative agreement is observed but the calculated curves are always below the experimental ones. The differences in the slopes of the numerical and experimental curves is mainly due to the deficiency of the retained hot-spot growth model and to uncertainties on the NM-temperature, which underestimate the energy-release rate and, therefore, the shock curvature.

Additional calculations have been conducted to model the experimental linear correlation between critical diameter and reciprocal of the GMB initial specific surface area (Figure 3). Such correlations exist for many heterogeneous explosives (Khasainov et al. [3], Presles et al. [7]). The most important prediction is that the calculations capture the shock sensitivity reversal effect. For monomodal mixtures (with GMB size either 5 μm or 47 μm or 102 μm), Figure 3 shows that the critical diameter is a non-monotonous function of the GMB size (Bouton [5]). The explanation is that the induction time of NM sensitized by the 47 μm and 102 μm GMBs is negligibly small compared to the burning time. The two corresponding lines are quite close to each other and to the universal empirical correlation, independent on the GMB size. Because of the imperfections of the retained critical-diameter model, we consider that the calculations fit reasonably the empirical line. On the contrary, the difference in the respective positions of the 5-μm line and of the two others is too large to be attributed to defects in the critical-diameter model. Indeed, the induction time for the 5-μm GMBs becomes comparable to the burning time, due to the relative increase of the conductive heat-dissipation rate compared to the viscoplastic heat-production rate around the GMBs.

For bimodal mixtures (with GMB sizes 5 μm and 102 μm), Figure 4 indicates that, at large detonation velocities, the shock curvature increases when the detonation-velocity deficit increases. At small detonation velocity deficit the calculated induction time around both 5 μm and 102 μm GMBs is small compared to the burning time. However, as the detonation velocity deficit increases, the induction time around the smaller GMBs increases more rapidly than that around the larger ones, and eventually becomes too large to induce ignition before the sonic locus. In this case, only larger 102 μm GMBs provoke the hot-spot growth so that the effective specific surface area of ignited hot spots is much smaller than the initial specific surface area of the considered NM-GMB mixture. This results in a D_n-C curve that exhibits two critical points. The part of the curve below the upper critical point is close to that associated with the 102-μm-GMB monomodal D_n-C curve (the slight difference is due to the fact that the NM volumetric fraction in the bimodal mixture is smaller than in the corresponding monomodal mixture). Thus, in case of bimodal distributions of size of potential hot spots (such as voids, GMBs or HE particles) one can meet the situation when smaller hot spots do not contribute to the chemical-reaction growth so that the critical detonation diameter nonmonotonously depends on the initial specific surface area of heterogeneous HE, as observed by Moulard [2].

Conclusions

Though the use of the detonation-shock dynamics model for calculating detonation critical diameters is questionable for the heterogeneous explosives considered here, because of their large (compared to the curvature radius) reaction zone lengths, a semi-quantitative agreement between experiments and calculations is obtained under the assumption of a viscoplastic GMB collapse mechanism in GMB-sensitized NM. However, this agreement cannot be considered as a validation of neither the assumed collapse mechanism nor of the critical diameter model. Yet the approach is useful because it defines correct orders of magnitude and the main trends and features of a complicated problem. This approach thus predicts the shock sensitivity reversal effect in mixtures of NM both with mono- and bimodal GMB distributions, but with significantly smaller size of finer GMBs than we could test experimentally. Such experiments are the subject of current efforts.

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