The Sensitisation of Thin Films of Nitroglycerine

G. D. COLEY* and J. E. FIELD
Surface Physics, Cavendish Laboratory, Free School Lane, Cambridge, England

High speed photography techniques have been used to study the sensitising effects of air bubbles (volume a few mm^3) within thin films of spark-ignited nitroglycerine. The collapse of the bubble is shown to lead to an increase in sensitivity of the liquid explosive, by (a) locally increasing the deflagration velocity and (b) generating a pressure pulse which is capable of producing hot spots at cavitation sites within the liquid. If several gas bubbles are included, cooperative effects are observed which lead to increased sensitiveness. The results support a distributed hot spot model for low velocity detonation.

Introduction

Low Velocity Detonation (LVD) in liquid explosive systems is a stable reaction regime, readily initiated by shock pressures of a few kilobars, with propagation velocities of the order of 2 mm/μsec. The thermohydrodynamic theory of detonation is not applicable to this regime since the pressures involved are too small for direct shock heating of homogeneous explosive to be significant. Recent experiments have shown that the presence of gas or vapour filled cavities within the explosive is an essential requirement for initiation and propagation of LVD. These inhomogeneities may either be present initially within the explosive, or they may arise as a consequence of precursor stress waves travelling through the confinement transmitting tensile forces to the liquid ahead of the reaction zone, thereby producing cavitation [1-4]. Thus, for the stable propagation of LVD in an initially homogeneous liquid explosive, there is a requirement that the stress waves must be able to propagate more rapidly through the confinement than the pressure waves through the explosive.

The ways by which bubbles sensitise liquid explosives are still not well understood although several sensitisation mechanisms have been identified. Bubble size has been found to be a particularly important parameter [4,5]. In the case of small vapour filled cavities there is strong evidence that adiabatic collapse of the bubbles, produced by compression shocks ahead of the deflagration front, forms hot-spots which facilitate the transfer to LVD. However, where large bubbles are concerned many factors could be operative in the sensitisation process. Bowden and McOnie [3] introduced a large air-bubble into a thin film of nitroglycerine (NG) confined by plates of PMMA and glass, and initiated the explosive by rapid spark discharge. They found that the deflagration products rapidly traversed the cavity giving rise to many new reaction sites on its far surface. We have noted similar effects with large air bubbles in thin films of nitromethane/nitric acid mixtures. Hay and Watson [6] injected a stream of gas filled bubbles into a three-dimensional configuration of an NG/ethylene glycol dinitrate mixture, and shocked the charge normally to the bubble stream. Initiation occurred in the centre of the stream, where bubbles were first collapsed, and reaction propagated preferentially along the stream. An interesting feature of their experiments was the formation of micro-Munroe jets in the bubbles, oriented in the direction of the initiating shock. They suggested that although jets may not directly initiate reaction, they may help it by the dispersion of liquid droplets within the bubbles.

The experiments described in this paper are

*A.W.R.E., Aldermaston, Berks.

Published by American Elsevier Publishing Company, Inc.

modifications of the thin film experiments previously described by Bowden and McOnie [3] and Coley and Field [4]. Air bubbles of controlled size were introduced at specific locations in the thin film situation, and their influence on the deflagration/LVD transition was examined. In previous experiments cavitation bubbles were produced in the liquid by precursor waves travelling at the liquid/confinement interface, and as a consequence there was very little control over their size and distribution. The deliberate insertion of air bubbles overcomes this difficulty, although the presence of air as well as explosive vapours does mean that there is not an exact equivalence between the artificial bubble field and a cavitation field. However, the presence of a gas bubble in a liquid explosive is clearly a practical situation.

This approach has yielded interesting information on the sensitisation of thin films of liquid explosives by large bubbles, and illustrated the cooperative behaviour between combinations of bubbles.

Experimental Details

Air bubbles of controlled size were introduced into a thin film of nitroglycerine confined between plates of PMMA. Deflagration was initiated in the film by spark discharge, and the interaction between the deflagration front and the bubble field was observed photographically with a Beckman and Whitley model 189 high speed framing camera operated at μsec frame intervals. A schematic diagram of the explosive round is given in Fig. 1. The film of nitroglycerine N was confined between two rectangular PMMA plates P_1 and P_2, size 25 mm X 80 mm, which were separated by two 0.5 mm spacers, S. The explosive was ignited by rapid condenser discharge across the spark gap formed by silver steel electrodes E_1 and E_2. The round was held rigidly together by two large clips B. C represents the air cavity introduced into the explosive film which because of its size was essentially cylindrical in shape.

In the initial experiments air bubbles were introduced into the explosive film by injecting air from a small diameter ($\sim$0.5 mm) hypodermic needle. This was later considered to be a hazardous method because of the possibility of the explosive becoming

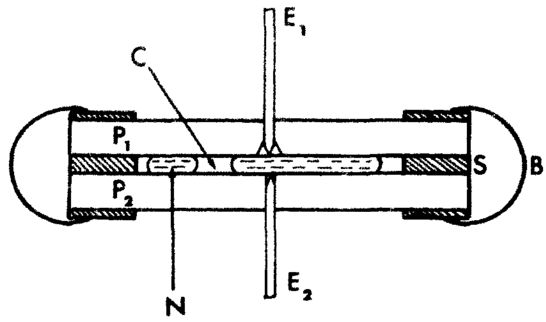

Fig. 1. Schematic diagram of the round. N is nitroglycerine, C gas cavity, P_1 and P_2 confining plates, E_1 and E_2 electrodes, S spacers, and B holding clips.

trapped in the needle, and initiating on compression; therefore, an alternative technique was developed. A length of fine copper wire, with a loop at one end, was inserted into the film, with the loop at the explosive air interface. By breaking the meniscus with the loop, it was possible to draw a small air-bubble into the film. Several air-bubbles were introduced by this method and then they were made to coalesce to produce one large air bubble of chosen size. Over a period of several minutes it was found that the bubble, particularly if it were relatively large, drifted towards the explosive/air boundary. To prevent this a piece of fine copper wire of diameter 0.2 mm was positioned on the far side of the bubble away from the spark source.

Experimental Results

In the sequence in Fig. 2 a large air-bubble of 3.7 mm^3 has been positioned 5.7 mm from the spark gap. In frame 1, deflagration has initiated, and a cavitation field is being produced by the action of precursor waves at the liquid/confinement interface. The bubble is clearly visible with its stabilising wire, and the explosive/air boundary can be seen in the top left corner of the frame. Between frames 1 and 19 the bubble is in a state of collapse. While the bubble collapse is accelerating there is a very marked increase in the deflagration velocity just in the region of the collapsing bubble (frames 14–20). Just before the deflagration front reaches the collapsed bubble a pressure pulse is produced, with a velocity of 820 m/sec, centred on the bubble.

Space/time plots for the reaction front in the collapse region, and remote from it, for the rapidly

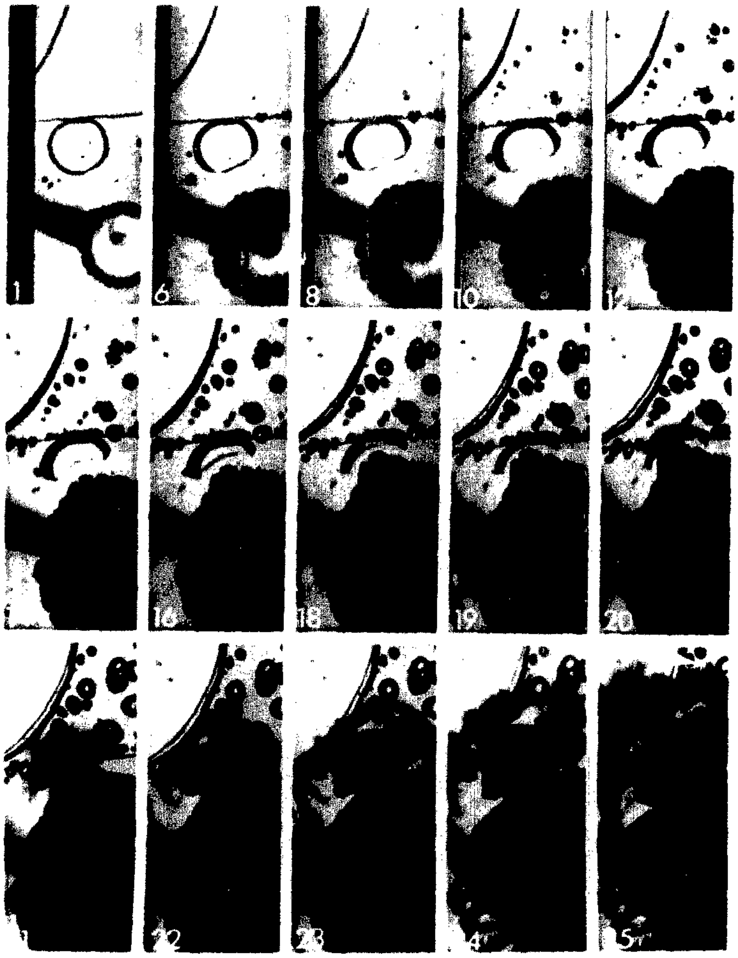

Fig. 2. Sensitising effect of an air bubble in a thin film of nitroglycerine and build up to LVD. Interframe time 1 μsec. Vertical extent of frame 16.6 mm.

collapsing wall of the air-bubble, and for the pressure wave, are presented in Fig. 3. A diagram showing the shape of the collapsing air-bubble at various intervals of time is also given as an insert to this figure. Initially the deflagration velocity is about 130 m/sec, but when the bubble achieves its maximum collapse rate of 300 m/sec the local deflagration velocity in the region of the collapsed cavity is 380 m/sec. As estimated from the graph the deflagration front impacts onto the rebounding bubble about 1.5 μsec after it has reached minimum volume. Reaction occurs at the collapsed bubble site as can be seen in frames 21-22. The radiated pressure wave collapses the cavitation bubbles that it encounters and reaction is produced at these sites within a μsec of the collapse. Hence, the reaction effectively keeps pace with the pressure front. Small ($<$ 0.04 mm diameter) cavitation bubbles are produced just ahead of the reaction front, and these are visible in the upper portion of frame 25 (arrowed).

In Fig. 4 a bubble of 4.6 mm^3 has been positioned 4.8 mm from the spark gap. The collapsing bubble wall reaches a maximum velocity of 470 m/sec. Initially the deflagration velocity is about 170 m/sec, but in the region of the collapsing bubble it accelerates to a final velocity of 690 m/sec; in regions remote from the bubble the front velocity is 290 m/sec at this time. The deflagration front reaches the bubble just as it achieves minimum volume and

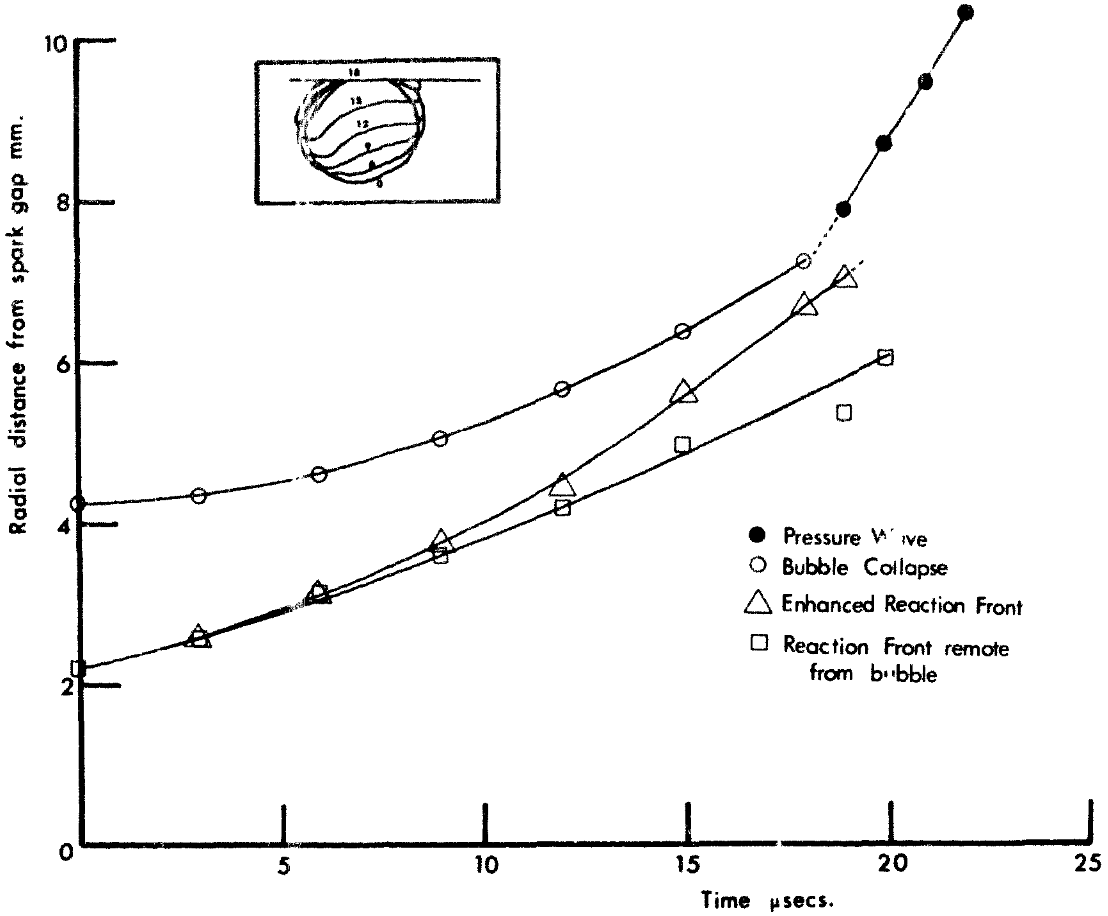

Fig. 3. Zone boundaries from Fig. 2. Insert shows bubble collapse as a function of time (µsec).

a shock radiates with a velocity of 945 m/sec. This shock collapses the cavitation bubbles and reaction develops at these sites.

A summary of the data obtained in experiments of this type is presented in Table 1. Average bubble diameters could be measured to ±2%; bubble volume and velocity values have an accuracy of about ±5%.

In some experiments several air-bubbles were introduced into the explosives film. Two bubbles were inserted in the sequence illustrated in Fig. 5. The larger bubble, volume 1.0 mm^3, was positioned 7.1 mm from the centre of the round, and the smaller bubble, volume 0.5 mm^3, 5.4 mm from the centre. Frame 2, in which deflagration has already initiated, shows the location of the bubbles relative

TABLE 1

Bubble Area mm^2	Bubble Volume mm^3	Distance between centres, Bubble/Electrodes mm	Initial Deflagration Velocity m/sec	Maximum Deflagration Velocity m/sec	Maximum Bubble Collapse Velocity m/sec	Shock Wave Velocity m/sec
9.3	4.6	4.8	170	690	470	945
7.4	3.7	5.7	130	380	310	820
3.6	1.8	4.5	120	530	350	980

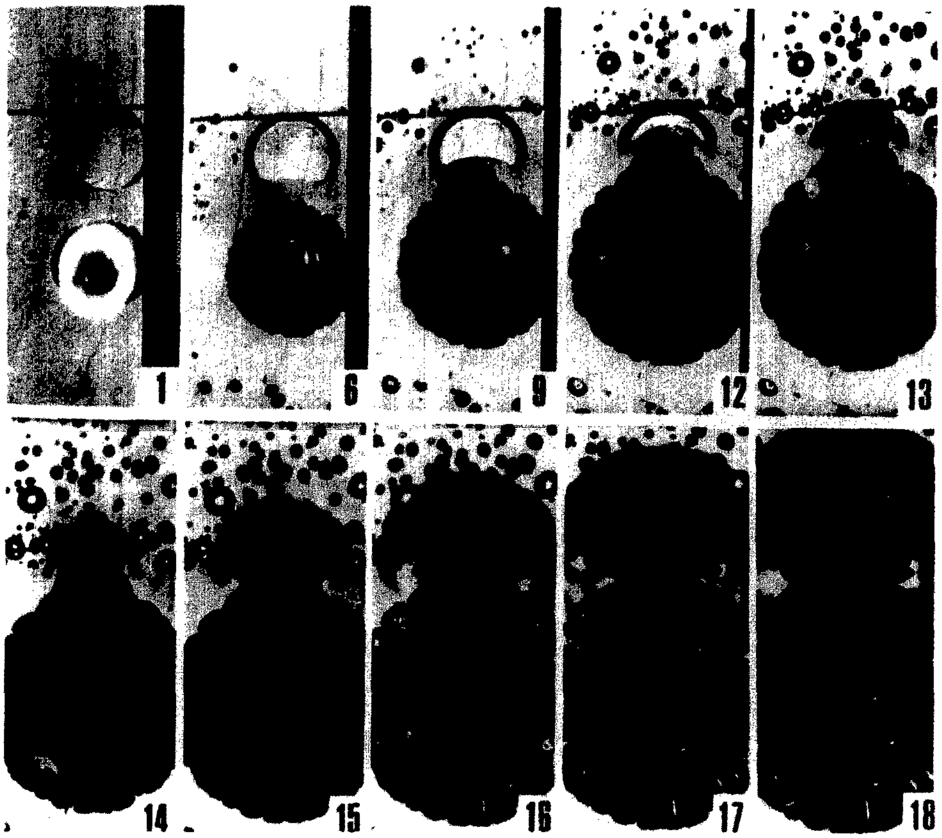

Fig. 4. Sensitising effect of an air bubble in a thin film of nitroglycerine and build up to LVD. Interframe time 1 μsec. Vertical extent of frame 16.5 mm.

to the spark gap. The small bubble is collapsed by compression shocks ahead of the deflagration, and reaches minimum volume between frames 14 and 15, and then rebounds. The large air-bubble is also collapsed by the shocks ahead of the deflagration front, but in addition to this, following frame 16, it is also influenced by the rebound shock from the small bubble. This particular shock produces a micro-Munroe jet within the large bubble, which is clearly visible from frame 20 onwards. The jet impacts with a velocity of about 120 m/sec on the far wall of the large bubble between frames 23 and 24 (not shown), but this impact does not produce any visible reaction. The deflagration front has accelerated to about 145 m/sec by frame 25, and up to this time the collapsing bubbles seem to have had little effect on the deflagration velocity.

In another experiment (Fig. 6) one large bubble, and two small bubbles, A and B, were introduced into the explosive. Bubble A initially expands under the action of the precursor waves at the liquid/confinement interface, and then starts to collapse from frame 8. Bubble B initially expands and is then collapsed by the action of shocks both from the deflagration front, and from nearby rebounding cavitation bubbles. After reaching minimum volume in frame 13 the bubble rebounds and jets towards the large air bubble; the jet moves with a velocity of $\sim$320/m/sec. In frames 22 and 23 the jet penetrates the wall of the large bubble and disperses droplets into it.

Discussion

If a large bubble is located very near the initiation source products tend to penetrate the bubble before

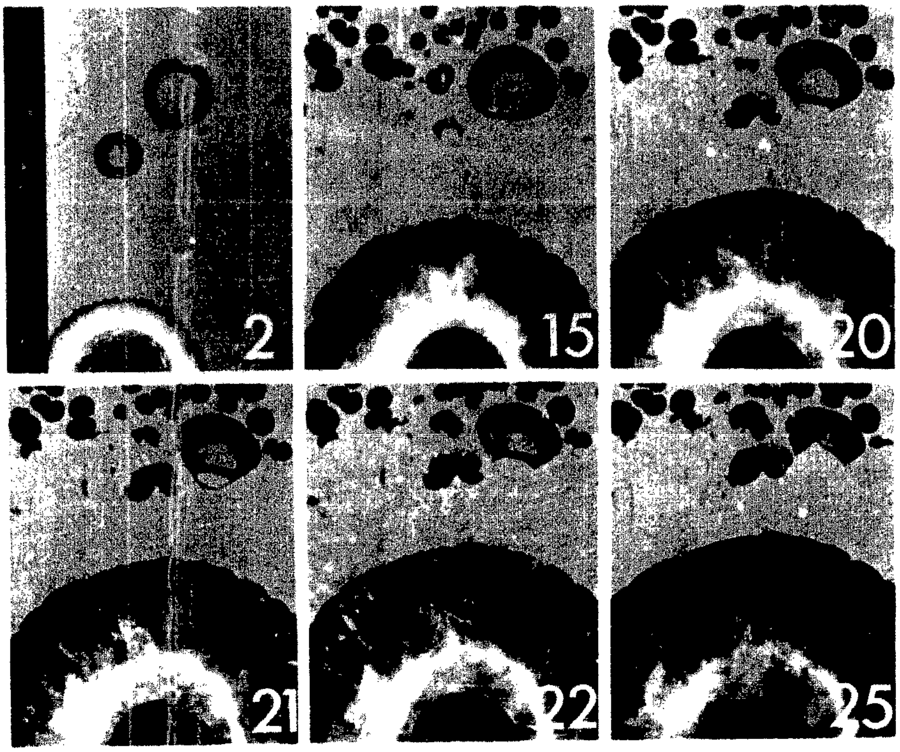

Fig. 5. Selected frames from a sequence in which two air bubbles were introduced into a film of nitroglycerine. Note the jet formation in the large bubble. Interframe time 1 μ sec. Vertical extent of frame 8.4 mm.

it has a chance to collapse. This process was illustrated by Bowden and McOnie [3]. Smaller bubbles which are collapsed very rapidly can cause initiation ahead of the main deflagration front [4]. This paper is concerned with bubbles of intermediate size at chosen positions relative to the initiation centre. All the experiments described here had certain general features. Compression shocks in the film ahead of the deflagration caused the bubble to collapse. During this collapse stage the deflagration front in the region of the bubble was rapidly accelerated towards it, whereas remote from the bubble the front accelerated at a much slower rate. The distance of the bubble from the centre of the round determined whether the deflagration front caught up with it before, at, or later than the minimum volume condition. In all these situations a shock wave was produced as a consequence of bubble collapse, and as it travelled through the film it collapsed cavitation bubbles producing adiabatic hot-spots.

In his treatment of the collapse of an empty spherical cavity situated in an ideal fluid, Rayleigh [7] predicted that infinite pressures would be generated on complete collapse. However, in the more realistic situation, in which compressibility and viscosity of the fluid are taken into account, and gases or vapours are allowed to be present within the cavity, large finite pressures result. In a paper by Jones and Edwards [8] minimum pressures of about 10 kbar have been cited for the collapse of vaporous cavities in water. Uncertainties about the minimum volume attained by the cavities in their experiments led them to believe that 10 kbar should be considered as a lower limit with the possibility of achieving around 100 kbar. Hay and Watson [6] suggest that since these higher pressures are similar to those required for the direct shock initiation of explosives [9], direct initiation of the explosive by the shock wave generated in the final stages of cavity collapse could furnish a fast reaction mechanism. They suggest, however, that vaporous rather than gaseous cavities would be more effective, since Hickling and Plesset [10] found that peak pressures reached during the final stages of the cavity collapse increased as the initial pressure within the cavity decreased. The bulk of the bubble collapse treatment has been concerned with the symmetrical collapse of spherical cavities [8]. However, in cases where bubble collapse is no longer symmetric very high pressures can result [11].

Brunton [12] has passed kbar strength shocks over disc-shaped air-bubbles in water trapped between two transparent plates and photographed the bubble collapse. Asymmetrical bubble collapse occurred in which the involution of the bubble wall produced a rapidly moving jet of liquid. Brunton recorded maximum speeds for the jet in the region of 500 m/sec. The impact of this high speed intrusion was responsible for a pressure wave in the liquid. Trapped gas within the bubble was important since this prevented the formation of a closed loop of liquid that would divide the disc-shaped cavity into two. Shock pressures of the order of 7 kbar would be generated by a 500 m/sec slug of water impacting on the far surface of the bubble [12]. In our experiments bubble collapse velocities of about 310, 350, and 470 m/sec were observed for the three different bubble sizes (see Table 1), and we would expect shock pressures in the region of a few kbars to be generated. These pressures are insufficient to produce shock initiation in homogeneous liquid explosives, and are also rather less than the 10 kbar usually associated with fast reaction. However, a shock wave of a few kbars propagating through cavitated liquid can produce initiation at other bubbles or cavity sites.

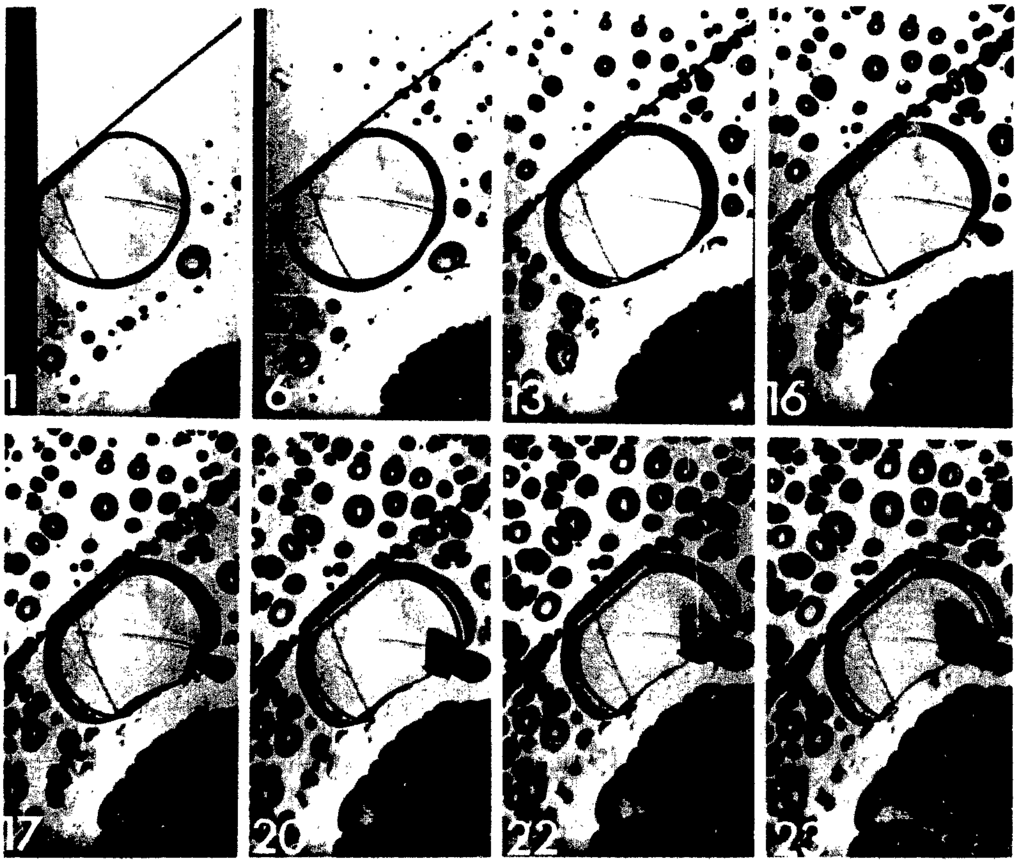

Fig. 6. Selected frames showing co-operative effect between air-bubbles. Note the way one bubble jets into the large bubble dispersing droplets into it. Interframe time 1 μsec. Vertical extent of frame 13.4 mm.

The sensitisation of the thin film situation by an intermediate size air-bubble appears to be as follows. The bubble collapse leads directly to an enhancement of the deflagration velocity, and also produces a pressure wave capable of adiabatically initiating nearby cavitation bubbles. Reaction can thus be initiated well ahead of the deflagration front by the pressure wave generated in the bubble collapse, and the reaction front effectively keeps pace with this pressure wave. Compression shocks from each of the reaction sites propagate through precompressed explosive and will therefore tend to overtake the leading pressure waves and form a pressure front. Thus we would expect the LVD to accelerate steadily to a stable velocity, being a shock wave controlled process, but depending on cavitation bubbles for the provision of reaction sites. A model similar to this was proposed by Schall [13], and the present work gives experimental support for this process.

When more than one air-bubble is introduced into the thin film situation interesting coupling effects occur between the bubbles. In the two bubble example, shown in Fig. 4, after reaching minimum volume, the small bubble rebounds, and the resulting shock wave causes a jet within the larger bubble. The rebound of the small bubble is facilitated by the fact that the larger bubble is collapsing under the effect of shocks ahead of the deflagration front. In an investigation of the interaction between two underwater explosion bubbles oscillating out of phase, Smith [14] showed that a jet could be developed in one by oscillations in the other. In similar manner a jet is produced in the large cavity of Fig. 6. This jet eventually breaks the bubble membrane and travels at ~ 320 m/sec into the interior of the large bubble. An interesting feature is the break up of the jet into droplets at its head which will be dispersed within the bubble. These droplets could participate in the reaction and significantly increase sensitivity as suggested by Johansson and Selberg [15].

Experiments for this film thickness, in which large air bubbles (volume a few mm^3) were not included, transfer to LVD at a later stage, when the burning front interacts with the much smaller cavitation bubbles. It appears then that gas bubbles can present an increased hazard by significantly affecting the sensitiveness of an explosive.

This work has formed part of a much wider programme in which the reaction in liquid explosives has been studied for various conditions of confinement and initiation, and it is hoped to publish these results shortly.

We thank A.W.R.E., Aldermaston, for a grant in support of this work and E.R.D.E., Waltham Abbey for the supplies of explosive.

References

1. **Watson, R. W., Summers, C. R., Gibson, F. C., and Van Dolah, R. W.**, *Fourth Symposium on Detonation* 4, 117 (1965).
2. **Dubovik, A. V., Voskoboinikov, I. M., and Bobolev, V. K.**, *Fiz. Goreniya i Vzryva* 4, 105–110, (1966).
3. **Bowden, F. P., and McOnie, M. P.**, *Proc. Roy. Soc. (London)* **A298**, 38–50 (1967).
4. **Coley, G. D., and Field, J. E.**, *9th International Congress on High Speed Photography*, Denver, Colorado (1970), p. 466.
5. **Coley, G. D.**, Initiation and Growth of Explosion in Liquids, Thesis, Univ. of Cambridge (1971).
6. **Hay, J. E., and Watson, R. W.**, Ann. N.Y. Acad. Sci. **152**, 621–635 (1968).
7. **Lord Rayleigh**, *Phil. Mag.*, 6th Series **34**, 94 (1917).
8. **Jones, I. R., and Edwards, D. H.**, *J. Fluid Mech.* 7, 596 (1960).
9. **Mader, C. L.**, *Phys. Fluids* 6, 375–381 (1963).
10. **Hickling, R., and Plesset, M. S.**, *Phys. Fluids* 7, 7–14 (1964).
11. **Kornfeld, M., and Suvorov, L.**, *J. Appl. Phys.* **15**, 495 (1944).
12. **Brunton, J. H.**, *Proc. 3rd. International Conference on Rain Erosion and Allied Phenomena* (Ed. A. A. Fyall), R. A. E., Farnborough, England (1970), p. 821.
13. **Schall, R.**, *Zeit. fur Angewandte Physik VI Band* **10**, 470 (1954).
14. **Smith, A. V.**, *Phil. Trans. Roy. Soc. Lond.* **A260**, 279 (1966).
15. **Johansson, C. H., and Selberg, H. L.**, *Appl. Sci. Res.* **A5**, 439 (1955).

(Received October, 1972; revised version received January, 1973)