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GROWTH OF BURNING TO DETONATION IN LIQUIDS AND SOLIDS

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(A) Summary of Work

An investigation is being made of deflagration and growth to detonation in liquids and solids with special reference to the role of small discontinuities in promoting sensitivity, and the Deflagration, Detonation Transition. As with previous reports it will be seen that the approach has been on a fairly broad front covering a range of experimental studies. The results are discussed under separate headings.

(i) Liquid Explosives

A study is in progress of the factors which affect the initiation and transition to detonation of explosion in liquids. In earlier reports and papers (see, for example, Bowden & McOnie, 1965, 1967) the importance of discontinuities such as bubbles of gas and cavities has been emphasised. Photographic sequences clearly showed that a slow burning could quickly transfer to a low velocity detonation when it encountered a cavitated region of explosive: the discontinuities were then able to sustain the rapid propagation of explosion. Related experiments indicated the importance of 'jetting' which was observed to occur in various situations, (i) during the collapse of a cavity, (ii) when a droplet with a curved surface impacts against another surface, (iii) when a distribution of droplets on an anvil are forced together by a falling hammer. In 'impact' (falling hammer) experiments the presence of grit particles in the film of explosive markedly affected the sensitivity of the explosive.

Experimentally much of the above work was performed on thin films of nitroglycerine confined between thin sheets of perspex or glass: initiation of burning was started at the centre of the film by firing a spark. Bubbles of chosen size were added to the film when required, but in fact cavitation phenomena were always present due to precursor waves. The photographic sequences were taken with a C_4 camera capable of an inter-frame time of 5 μ secs.

Recent work has extended several aspects of the above:

(a) A detailed investigation is being made of the collapse of cavities by shock using high speed photography. It has been shown that when a shock of strength about 1 Kbar collapses a millimeter sized cavity, there is a rapid decrease in volume (to about $^1/_{60}$th of the initial size) and a temperature rise to about 1500^{o}K. During the collapse involution of the cavity occurs and a jet is formed which crosses the cavity first impacting and then penetrating the far side. This jet may reach velocities of a few hundred metres/sec. Following the

collapse there is a rapid expansion which sends a shock into the liquid. Both this shock and the jet impact can cause damage to a neighbouring solid. Clearly these three processes have importance in various explosive situations.

(b) Inert liquids (example, water) have been used in the thin film confinment as used in the earlier work. This has been useful in assessing various features of the cavitation process. For example, with perspex confining plates:

(i) the cavitation front moves typically at 700 metres/sec although values as low as 440 metres/sec and as high as 1050 metres/sec are observed.

(ii) the size and density of packing of the bubbles seems to depend on the thickness of the confinement and the spark energy. Thin layer confinement leads to densely packed bubbles growing to large sizes.

(iii) the cavitation bubbles do not visibly move in position during their initiation, growth and collapse.

(iv) in all experiments, after some time has passed, the cavitation bubbles which have collapsed rebound and then oscillate. At this stage they are usually elongated along a radial direction: this is consistent with the 'jetting' which was observed in the more large-scale experiments on single drops discussed above. (Remember that these experiments are with inert liquids, in the explosive situation the cavities are collapsed by compression shocks from the burning front, there is then no oscillation. The transition to detonation occurs when the burning accelerates sufficiently to reach cavities before they are collapsed.)

(v) the cavitated region always starts some distance out from the centre of the film, about 10 - 15 μ secs after the spark has fired. It is thought that the liquid in the immediate vicinity of the spark is too compressed for the tensile precursor waves to initiate cavitation.

(vi) by choosing suitable geometries it has been possible to demonstrate that cavitation is not associated with reflected waves either from the liquid or solid surfaces.

The evidence at present is consistent with the view that of the stress waves generated during the spark discharge (and the burning in the explosive situation) it is the Rayleigh surface wave which is responsible for the cavitation.

(c) The experiments on thin films of nitroglycerine have been photographed recently at much higher framing rates and magnifications using a Beckman and Whitley (model 189) rotating mirror camera. (This camera is capable of a framing interval down to 0.25 μ secs compared with the 5 μ secs of the C_4 camera with which the earlier work was performed.) It has been possible to

follow the detailed interaction of the burning front with the cavities and to study retonation waves back through the burning and the low velocity detonation wave passing through the cavities.

The sequence of behaviour when a thin film of nitroglycerine is confined between plates and spark initiated is, therefore, as follows:

(i) the spark initiates burning and sends stress waves through the liquid and the confining plates. Of the stress waves in the plates the Rayleigh surface wave has the largest surface displacement and causes cavitation in the liquid. The surface displacements in the plates do not return to zero because the pressures from first the spark and then the burning force the plates apart.

(ii) the cavitation front, when there is perspex confinement, moves through the liquid with a velocity of about 1000 metres/sec, the burning follows at a lower speed of approximately 300 metres/sec.

(iii) compressive shocks from the burning tend to close the cavities ahead of them, but as the burning accelerates the gap between the burning front and the rear of the cavitation closes.

(iv) a sudden transition to a low velocity detonation occurs when the burning front enters cavitated liquid.

(v) this low velocity detonation moves at a velocity of about 1200 metres/second through the liquid, while at the same time a retonation front returns back through the deflagration region.

Bowden, F.P. & McOnie M.P., Nature, 206, 4982, 380-3, 1965.
Bowden, F.P. & McOnie, M.P., Proc.Roy.Soc. A 298, 38-50, 1967.

(ii) Solid Explosives: initiation and propagation of explosion

Much of the recent work has been with single crystals of the azides (silver, lead, sodium, thallium) and with P.E.T.N. and has been an extension of earlier work on the effect of discontinuities on explosion. Various types of experiment have been performed.

(a) single crystals of silver and lead were mounted in a water bath with a small air bubble attached to the crystal. Collapse of this bubble by a weak shock of strength about 1 Kbar caused initiation of explosion. The various processes which occur when a cavity is collapsed have been discussed above. By careful choice of geometry (i.e., by varying the position of the cavity and the direction of the shock) it was possible to distinguish between the three possible initiation mechanisms (i.e., adiabatic heating, jet impact, expansion shock), and to demonstrate that the major effect is the high temperature produced in the cavity

during collapse. The photographic records allowed the volume changes of the cavity to be measured and hence the temperatures to be calculated (by assuming adiabatic conditions). The photographs also provided an estimate of the contact area between the cavity and the crystal and showed that the initiation took place within 1 μsec of the cavity reaching minimum volume. An account of some of these experiments has recently been published (Bowden & Chaudhri 1968).

(b) a study has been made of the rates of propagation of chemical reaction in lead azide. The velocities were found to depend on the minimum dimension of the crystals: values in the range 1000 to 2700 metres/sec were recorded. A special feature of this study was the examination at high magnification of the zone ahead of the flame. When the reaction travels at about 1000-1500 metres/sec, the stress waves in the crystal move ahead and cause mechanical effects such as fracture and disintegration of the crystal. However, though it seems more than possible that this fragmentation aids the propagation (see below under the section on decomposition) we have never found secondary initiation ahead of the flame. The behaviour of silver azide was found to be very similar to that of lead azide.

(c) the initiation of explosion in lead azide was investigated by bringing crystals into a hot furnace. The temperature for explosion depended on crystal size. By comparing the results with thermal explosion theory it is concluded that in the initiation of explosion in this manner some regions are formed which have a higher rate of decomposition than the normal crystal surface. It is at these locations that explosion starts. Defect structures in lead azide are currently being investigated, and it is hoped that it will be possible to relate areas of preferential decomposition with particular types of defect.

Bowden, F.P. & Chaudhri, M.M. Nature, 220, 5168, 690-4, 1968.

(iii) Solid Explosives: decomposition

As discussed in earlier reports and papers (see, for example, Walker, Gane and Bowden 1966) studies have been made of the decomposition of the series of azides sodium, thallium and lead. In these experiments measurements were made on individual single crystals, thus eliminating many of the complications inherent in studies of crystalline compacts such as ill-defined size distributions and packing factors. Methods were developed for growing crystals of chosen uniform size (see Walker et al. 1966). The smallest crystals produced had diameters of about 1000 Å while the largest had millimetre dimensions. The kinetics of decomposition were measured using ultra high vacuum techniques.

One of the main conclusions from this work was that the rate of decomposition was in all cases directly proportional to the area of the reacting surface. The conclusion was substantial over a wide temperature range (for example, 100^{o} to 450^{o}C for lead azide).

Studies with a scanning electron microscope showed that decomposition proceeds along specific crystallographic planes. Metal produced in the reaction did not appear to play an important part in the kinetics of decomposition.

More recently experiments have been in progress for measuring the decomposition which occurs when a crystal is cleaved. This involves cleaving crystals in a high vacuum apparatus and in the presence of a mass spectrometer so that any decomposition can be monitored. A wide range of crystals were used ranging from thermally very stable crystals such as MgO, through a range of carbonates (those of calcium, magnesium and lead) of decreasing stability to explosively unstable materials such as lead and sodium azide and P. E. T. N. With the exception of magnesium oxide all the crystals gave detectable thermal dissociation products. The amount of decomposition was found to depend on both the crystal type and the speed of fracture. Expressed as an average number of monolayers decomposed the amount varied from a fraction of a monolayer to a few monolayers. In the case of β-lead azide 10 monolayers were involved and it seems possible that partial initiation took place.

If dynamic crack growth is considered the strain energy can be regarded as dissipated in various ways; the energy to produce new surfaces (elastic surface energy), kinetic energy given to the specimen, plastic processes. Our interpretation of the decomposition is that it is localised at the moving crack tip. The atoms in this region will clearly be vibrating with large amplitudes because of the rupture process. A distribution of amplitudes would be expected because of the 'background' thermal vibrations always present. Atoms which dissociated would be those in which the combined thermal and 'rupture' vibrations added sufficiently to break bonds.

It is possible knowing the velocity of the crack and the amount of decomposition to work out a rate of dissociation. If the kinetics of decomposition for a particular crystal are measured separately it is possible to use the standard Arrhenius type equation to work out an 'effective' temperature at the crack tip. This procedure has been followed for all the crystals mentioned above. With lead azide the 'effective' temperature at the crack tip for a fast moving crack was about 570^{o}C.

The fact that decomposition is associated with fracture is an important result. It may be that under certain circumstances initiation of an explosive may be started by a fracture mechanism. We have already mentioned above that fractures often run ahead of a burning front. The production of local 'hot spots' at the crack tips or of pockets of gas which could later be compressed adiabatically could accelerate the burning process or in the case of a low velocity detonation sustain it.

A short note on the direct observation of thermal decomposition produced by fracture has recently been published (Bowden, F.P., Fox, P.G. & Soria-Ruiz, J. Nature, 220, 5169, 778-9, 1968).

Inert Solids

A programme of work is in progress in the laboratory for studying fracture growth in solids. Many of the results provide useful information for certain aspects of the explosives work. The results of this study can be briefly summarised as follows:

(i) In brittle solids cracks can quickly accelerate to high speeds. Velocities of the order of 1000 metres/sec can be reached in times of the order of a microsecond.

(ii) For many brittle solids a maximum velocity of crack propagation exists. This velocity can be very high (1500 metres/sec in soda glass, in excess of 5000 metres/sec for magnesium oxide).

(iii) The maximum speed in essentially isotropic solids is limited by crack branching which occurs for a given solid at a critical velocity. If attempts are made to drive single cracks at higher speeds than the critical branching velocity repeated crack forking ensues. The critical velocity (and hence the maximum velocity) is usually about 0.3 of the dilatational wave velocity or in terms of the Rayleigh surface wave velocity about 0.5.

(iv) In solids with well defined cleavage planes crack branching is suppressed until much higher speeds, and maximum velocities much nearer the Rayleigh wave velocity are achieved (i.e., about 0.6 of the dilatational wave velocity).

(v) Crack motion can be strongly affected by stress waves. The interaction may even cause crack branching.

(vi) Very intense stress waves can cause initiation of many individual cracks along their path. In these circumstances a fracture front can travel at the stress wave speed even though the individual cracks travel somewhat slower.

For further details of the fracture work see,

Bowden, F.P. & Field, J.E. Nature 214, 5110, 38-41, 1967.

Field, J.E. & Heyes, A.D. Proceedings 7th Int. Congress on High-Speed Photography, Zurich, 1965, p.391.

(B) Difficulties Encountered

No major difficulties have been encountered in the period covered by this report.

(C) Research Plans

Much of the proposed work has been indicated in the earlier part of this report. New studies will include (i) Liquids: a continuation at higher framing speeds and magnifications of the deflagration detonation transition in nitroglycerine. An extension to other liquids, different confinements and a 3-d situation. (ii) Solids: a continuation of the work on azides and P.E.T.N., and the factors which affect sensitivity; a closer study of defect structures in these explosive materials and the relation between reactivity and solid state properties; a better assessment of the role played by fracture in initiating (?), sustaining explosion; a particular problem exists in explaining the relative sensitivities of lead and sodium azide - the former is much the more sensitive but its rate of slow thermal decomposition is appreciably lower. It is possible that the relative strength and fracture properties are important factors in explaining this apparent paradox, and this is being investigated; use of an image converter camera system and a transient light detector to investigate 'spontaneous' explosion in lead azide; initiation by ultra-violet light, laser light and micro particle impact.

(D) Inventions

No inventions have been made during the period of the grant.

(E) Personnel

The work performed under this contract has been supervised by Dr. D. Tabor, Dr. J. E. Field and Dr. A. D. Yoffe.

Dr. P. G. Fox is studying the decomposition of the azides using the ultra high vacuum and mass spectrometry techniques. M. M. Chaudhri is currently studying the initiation of fast decomposition in explosive crystals. Dr. T. J. Bastow, M. J. Twigg and R. E. Winter are concerned with laser, fracture and micro-particle impact work, respectively. G. D. Coley who is investigating the deflagration detonation transition in liquids is supported by A.W.R.E. Aldermaston, who finance in part work on liquid explosives in the laboratory.

(F) Conferences, Travel

Dr. J. E. Field attended and presented a paper at the 2nd International Congress on Fracture, Brighton, April 13th - 18th, 1968.

M. M. Chaudhri will participate in the 5th AFOSR Contractors' Meeting on 'Combustion Dynamics' in Denver, Colorado, 23rd - 27th June. He will present a paper on some recent work under this contract.

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13. ABSTRACT

An investigation is being made of deflagration and growth to detonation in liquids and solids with special reference to the role of small discontinuities in promoting sensitivity, and the Deflagration, Detonation Transition. As with previous reports it will be seen that the approach has been on a fairly broad front covering a range of experimental studies. The results are discussed under separate headings.

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