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FINE STRUCTURE IN NITROMETHANE/ACETONE DETONATIONS

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Dark waves in the detonation front of nitromethane/acetone mixtures, known for 30 years, have been photographed using pairs of image intensifier cameras, triggered at slightly different times. The photographs show the motions and changes of the complex wave structure. Intriguing results are that the average brightness doesn't change as much from center to edge as simple theory would predict, and brightness goes down rather than up when a reflected shock from a glass plate enters the explosive products. The structure remains visible in the products stagnated against a glass plate, showing that the bright regions are high temperature regions, not just areas where less absorbing material is present ahead of the emitting layer.

The purpose of this paper is to present some new photographs of the luminous detonation front of nitromethane and mixtures of nitromethane and acetone. The photographs show some details of the fine structure apparently caused by transverse waves in the detonation reaction zone.

Studies of the transverse wave effects are not new. The waves were first observed by Campbell, et al. (1), in 1956, and have been the subject of many papers. There were contributions to the last Detonation Symposium by Kato and Brochet (2) and Persson and Persson (3), and to the Sixth Colloquium on Gas Dynamics by Dremin and Persson (4), devoted to the luminosity of the front. Mallory (5) has studied the pressure variations in the detonation front by photographing the development of roughness in a reflecting surface driven by the detonation. The structure of the detonation front in gases and in condensed phase explosives has been reviewed by Fickett and Davis (6). An interested reader can find most of the pertinent work from the reference lists of these sources.

The work described here and most of the previous work employ detonations in cylindrical tubes, and study the detonation after it has propagated far enough to become steady in its average motion. The fluid mechanics of a laminar reactive flow forming a steady detonation in a cylindrical tube has been studied for a long time, most recently by Bdzil (7,8). Figure 1 is a diagram showing the main features of the detonation reaction zone. The leading element is a curved shock wave; the resulting compression heats the material and initiates the chemical reactions. At the center, the shock pressure is high, nearly that of a steady, plane detonation. At the edge it is appreciably lower and thus curved back, as it matches into the flow in the inert confinement. The reaction takes longer in the lower temperature region near the edge than it does at the center, as indicated by the dashed line marked complete reaction. Between the shock and the complete reaction surface is the sonic locus. Only energy released between the shock and the sonic locus can propagate forward to drive the detonation wave. Along the central streamline the pressure jumps to a high value at the shock, and then decreases as reaction proceeds, much like the ZND plane detonation. Along a streamline away from the center the sequence of events is qualitatively much the same, but the pressures are lower so the reaction takes longer, and there is a transverse component of the velocity, varying as reaction proceeds. Little is known about the equations of state for the explosive and its products, but most workers believe that the temperature jumps to a high value, say between 2500 and 3000 K, at the shock, and then rises another few hundred degrees as reaction takes place and the pressure drops. The temperature at the shock is expected to be a few hundred degrees lower at the edge than at the center. The pictures presented here are of the light emitted from the detonation front through the transparent unreacted explosive ahead. Not enough is known about the opacities of the materials, and even of the spatial distribution of reaction, to allow calculation of the intensity expected from a reasonable laminar model.

The light emitted from the detonation front, especially from nitromethane diluted with acetone, is not very bright, and the light gain available with an image intensifier camera (9) is needed to make sufficiently short exposures possible. The photographs were taken with a

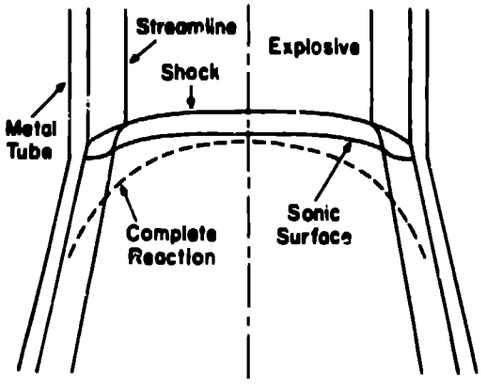

Fig. 1. Diagram showing the features of a detonation in a metal tube.

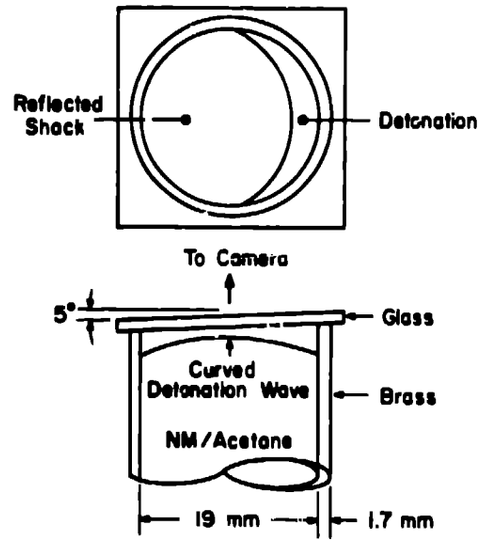

Fig. 2. Diagram of the experimental arrangement used to obtain photographs of the detonation front and of detonation products subjected to a reflected shock.

40-mm-diameter image intensifier camera with exposure time set to about 30 ns, and gain in the multiplier stage chosen for the particular dilution. The explosive was in a brass tube, 19-mm-inside diameter, with 1.7-mm wall. The tube was about 180 mm long. Initiation was accomplished with a small plane wave lens and 25-mm cube/or cylinder of Composition B. The downstream end of the tube was cut at a 5° angle, and covered with a glass plate. A diagram is shown in Fig. 2. The exposure was timed to photograph the detonation wave intersecting the plate. The curved wave intersects the plane plate in a curved line; on one side of this line the unperturbed detonation is seen, and on the other side the reacted detonation products are seen, after they have been compressed by the shock reflected into them from the glass. Reflection from glass increases the pressure about 30%. The phase velocity of the intersection of the wave with the glass is about csc 5° = 11 5 times the detonation velocity, so the intersection line moves about 2 mm during the exposure and is smeared by that amount. Two cameras were available for most of the photographs, and they were arranged to view the same subject through a beam splitter. The exposure times were offset to give views of the subject at two different times.

Photographs of the luminous detonation front in pure nitromethane are shown in Fig. 3. The front has intersected the glass plate at the left hand side of the circular luminous area. The intersection line is fuzzy because it moves during the exposure. There are two qualitative features to notice. First, the brightness does not decrease appreciably between the center and the edge. Other studies have shown, using other instruments, the same thing. The laminar flow model would predict appreciable change. Second, the region where light is emitted from material that has been additionally compressed by the reflected shock is less bright than the detonation front. Although the average temperature must have increased, the brightness has decreased.

Careful photometry has not been done, but perhaps some estimates of the brightness temperatures may be useful. The brightness temperature for the nitromethane detonation has been measured (10) before as about 3380 K. The region of the reflected shock has a brightness temperature of about 3120 K.

Photographs of the luminous front in nitromethane diluted with 15% acetone, Fig. 4, show that the real flow is not laminar and steady, but has an elaborate structure. In the detonation region the structure changes, and it is difficult or impossible to find correlations between the two exposures. In the reflected shock region, on the other hand, there is no change, and the structures seem identical in the two pictures. There is no appreciable change in either type of structure between the middle and the edge. The brightness variation is greater in the detonation region than in the reflected shock region. The calibration of the system is not adequate for obtaining good absolute brightness temperatures, but the differences can be evaluated reasonably well. The resolved variations in the detonation region lead to estimates of 2675 K at the bright areas, and 2450 K in the dimmest areas. The corresponding

Fig 3. The luminous detonation front in pure nitromethane, photographed at two times. At the left of the circular luminous area the front has intersected the glass plate, and the lunate area at the right is light from undisturbed detonation front. The dividing line moves across the frame about one tenth of the diameter during the exposure. The bright spots at the left are caused by bubbles left under the glass. C-4980-B&C.

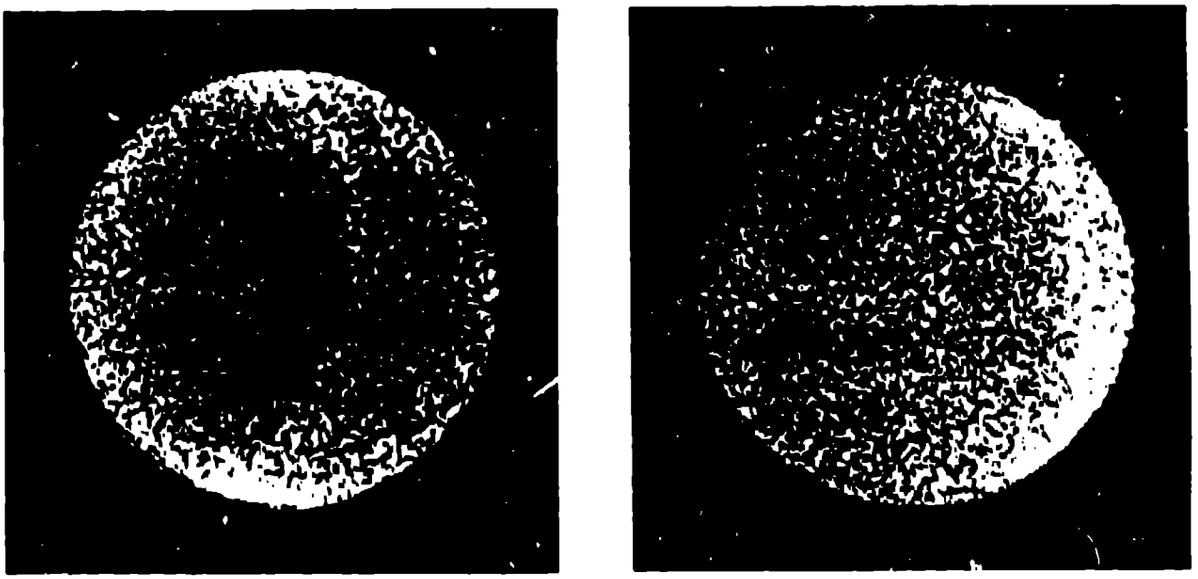

Fig 4. Photographs similar to Fig. 3, but using nitromethane diluted 15% with acetone. In the later frame the detonation has almost run off the edge at the right. Note that the structure in the reflected shock region, at the left of each frame, does not change with time. C-4948-C&B.

brightness temperatures in the reflected shock region are 2510 K and 2370 K, a somewhat smaller fractional change. These estimates certainly do not give the full variation, because the exposure time is too long to resolve small bright areas that are moving or changing, and light is scattered into the dark areas.

The size of the fine structure depends on the dilution ratio. Figure 5 shows photographs of the luminous front for nitromethane/acetone 90/10 and 80/20. Other dilutents, for example toluene, have also been used, with similar results. The scale of the structure is probably in direct proportion to the chemical reaction time, and the time is lengthened by dilution. It seems likely that structure occurs in all nitromethane detonations, including that of the pure material, and probably also in many other liquid explosives. If cameras with shorter exposure times become available in the future, perhaps direct observation of structure in pure nitromethane will be possible.

Looking at Figs. 4 and 5 it is obvious that the light is produced in the local regions of the structure, and does not seem to be influenced much by the curvature of the surface. The edge doesn't differ from the center. If detonation in the pure material also has structure, then its detonation is also local, and one can understand why the brightness doesn't decrease at the edge. Although Fig. 1 must be correct in some average sense, it does not model the details of the chemical reaction properly.

The decrease in brightness temperature in the reflected shock region compared with the detonation front can also be understood if the front has structure. The detectors used for photography, in our case the image intensifier cathode, are very nonlinear thermometers. The cathode used for these photographs responds, in the region of interest, approximately as the eighth power of the temperature. Therefore, if a small fraction of the area of the detonation front is at a much higher temperature than the average, and the small areas are not resolved in the photographs, it will appear that the brightness temperature is higher than the average temperature. The observation seems to indicate that there are small regions of a detonation front that are much brighter than the average, and that these small regions cool quickly and disappear after the detonation front reflects from the glass.

Although the measured temperature variation in the detonation region is larger than that measured in the reflected shock region, this variation is not enough to explain the observation. Suppose that the area radiating at a given brightness temperature is described by a normal distribution, and that the detector response varies as the eighth power of the temperature. If the standard deviation is 0.2 times the mean temperature, the apparent brightness temperature will be only 1.12 times the mean temperature. The real distribution must have a population of small areas at very high temperature. Perhaps these correspond in some

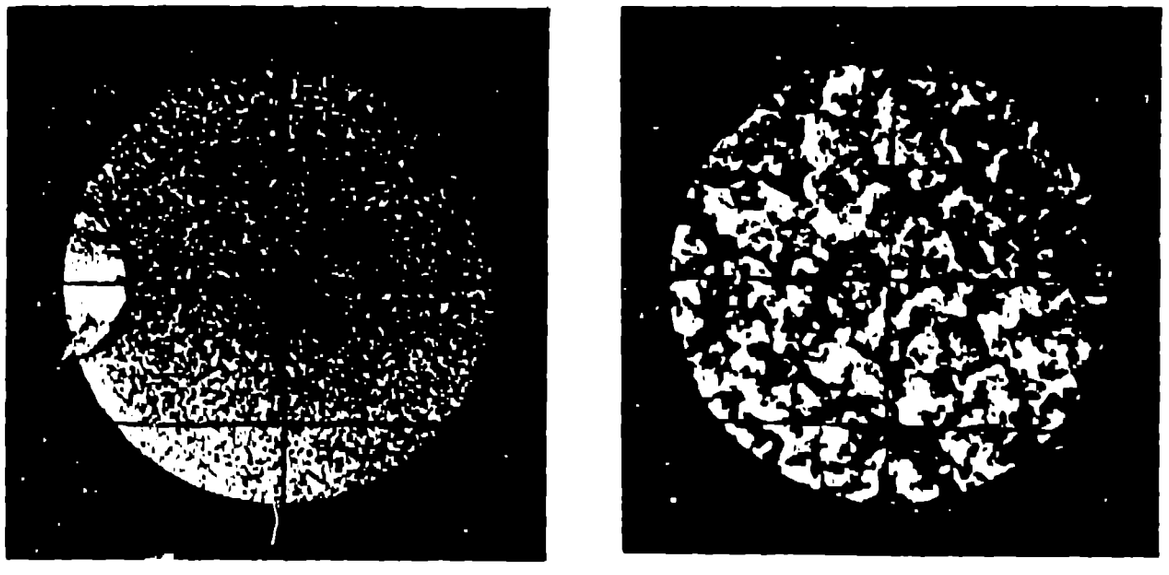

Fig. 5. Photographs of the luminous front for nitromethane/acetone 90/10 and 80/20. The size of the structure changes with the dilution. The perturbation at the left is caused by a trigger switch. The wires were intersected by the wave in the left picture, and waves proceed in each direction. C-4039-B and C-4842-B.

way to the observed Mach interaction regions in gas detonations.

In discussing apparent brightness temperature, we have assumed that the pictures can be interpreted in the simplest way, and that what we think we see is what is really there. One possible problem might be that the glass surface is damaged at the instant of reflection of the wave, and that it does not transmit all the light. A few experiments with the glass replaced by sapphire and with lithium fluoride show brightnesses that seem consistent with the glass, but the shock strengths are different and the corrections are doubtful. The experiments presented show that the apparent brightness doesn't change as the shock proceeds through the glass, so the shock doesn't make the glass absorbing. Other experiments with embedded mirrors confirm this finding.

Although the main aim of this paper is a discussion of light emission, some other things came up during the experimental program. One of these bears on the question of whether the structure on the detonation front is directly connected with that observed from the side. Fickett and Davis (6) suggested that the patterns observed by others (3,4) resulted from reaction in a ring behind the sonic surface, and were not observed from the front. Figure 6 shows the luminous front in pure nitromethane after the whole of it has reflected from the glass plate. Some waves from the edge can be seen as fine curved lines. Perhaps these can be related to the waves observed in open camera photographs of detonations in glass tubes. Pictures of both kinds for the same experiment will be needed to find out. (The pictures shown here in Fig. 6 were not obtained intentionally for the purpose; they were obtained when the timing was set incorrectly).

A puzzling observation came when we tried to do these experiments using nitromethane diluted with 20% acetone. The structure is larger and the pictures are better. However, the detonation front does not have the shape shown in Fig. 1. Instead, the edge leads the center and the wave is concave. Figure 7 shows a photograph more or less like the ones in Fig. 4, but the intersection of the wave with the glass is curved the other way. The wave velocity is about 5.5 mm/μs, well above that of any expected wave in the brass tube. We have no idea how to interpret this observation.

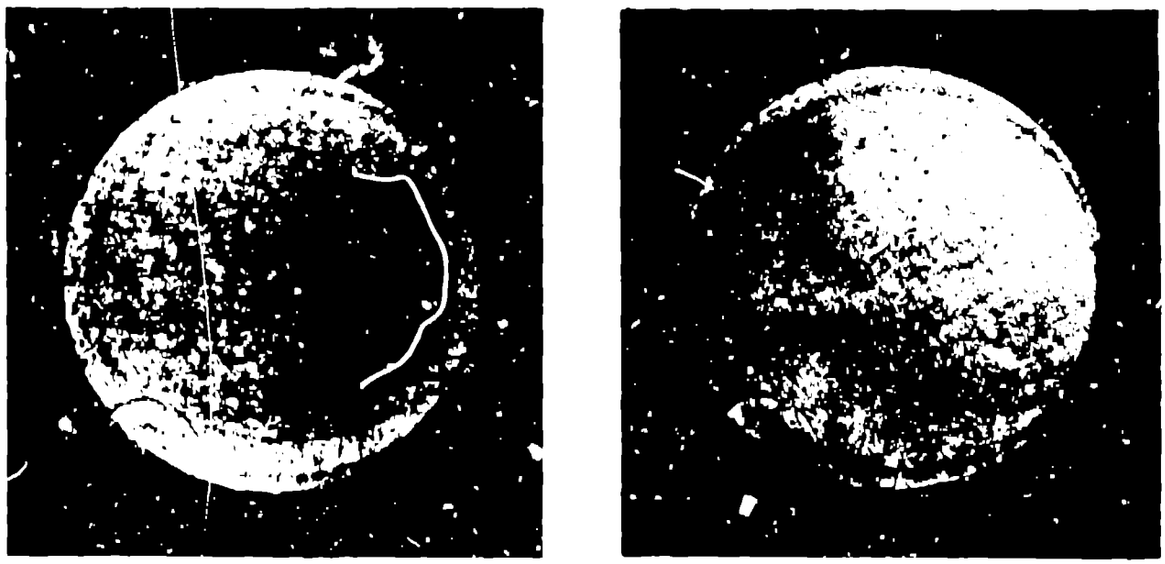

Fig. 6. Late photograph of nitromethane detonation, showing the luminosity after the whole wave front has intersected the glass plate. The waves around the edge may be caused by failure waves occurring in the slowly reacting explosive in the flow near the tube wall, as shown in Fig. 1. C-4949-C&B.

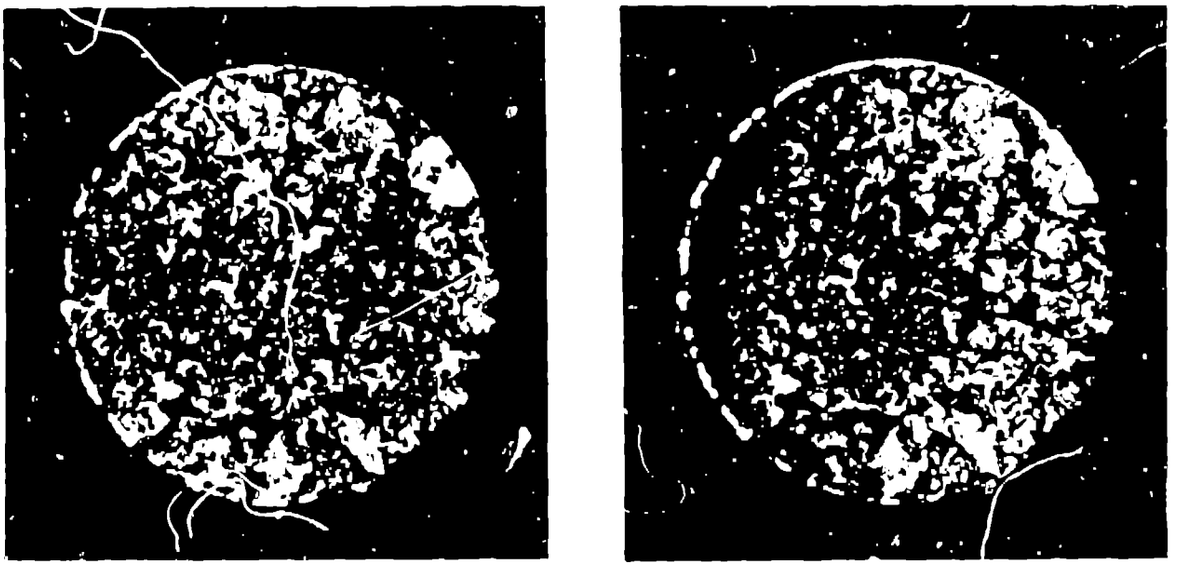

Fig. 7. Photographs similar to Figs. 3 and 4, but using nitromethane diluted with 20% acetone. The wave is not convex but concave, and intersects the glass at the edge rather than at the middle. C-4945-B&A.

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