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TITLE: HUGONIOT MEASUREMENTS IN BROMOFORM: LOOKING FOR SHOCK-INDUCED REACTION

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HUGONIOT MEASUREMENTS IN BROMOFORM: LOOKING FOR SHOCK-INDUCED REACTION†

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ABSTRACT

Electromagnetic particle velocity gauging has been used to measure shocks in bromoform to determine at what pressure shock-induced reaction occurs. Earlier work done at Los Alamos seemed to indicate reactions may occur at pressures as low as 3 GPa. Five experiments were done between 3 and 10 GPa. No evidence of reaction was observed in any of these experiments, although the data at 10 GPa had some unexplained perturbations in them. The data were very close to the universal liquid Hugoniot prediction for bromoform. Based on this and the earlier higher pressure data, it is estimated that reaction begins at shock pressures above 10 GPa, eliminating this material as a candidate for further easy study on our single-stage gun.

I. INTRODUCTION

Bromoform ($CHBr_3$) shock experiments have been carried out at Los Alamos in the past by Ramsay[1] in the early 1960's and McQueen[2] in the early 1980's. Motivation for the early experiments centered around understanding why some explosive liquids become opaque during the initiation process. Nonexplosive liquids were studied in an effort to find a material that goes opaque under shock conditions. Bromoform was found to exhibit this characteristic above 6 GPa with induction times of 1 to 2 μs.[1] Although Hugoniot measurements were made from 3 to 24 GPa, no definite reason for the material becoming opaque was determined. It was noted, however, that the Hugoniot data appeared to have an odd shape when compared with the water Hugoniot in the shock-velocity particle-velocity plane. In the later experiments McQueen observed that when bromoform is shocked to higher pressures (above 20 to 30 GPa), the shock front emits radiation that is sensitive to the shock pressure. In fact this material is being used by McQueen[2] as both a shock time-of-arrival detector and an indicator of wave profile changes occurring in materials in contact with the bromoform (by measuring the emitted light-time profiles).

The above information led us to look at bromoform as a possible material for further study, to determine if the reactions (particularly the low pressure reactions) were measurable with particle velocity gauges. Because of its high density, 2.89 g/cm^3, pressures up to 10 GPa could be obtained in single-shock experiments using the single-stage gun available to us. Based on recent work we have done using the "universal" liquid Hugoniot developed by Woolfolk, Cowperthwaite, and Shaw[3] at SRI to model the behavior of many liquids (and establish evidence of shock-induced reaction based on deviation of the Hugoniot data from it),[4] it appeared that bromoform might exhibit reactive behavior even below 3 GPa. With this in mind, five electromagnetic particle velocity

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gauging experiments were conducted on bromoform in an effort to measure evidence of the reactions.

II. EXPERIMENTAL SETUP

The experiments consisted of a liquid cell 68.6 mm in outside diameter made from Kel-F plastic; a 3-mm-thick Kel-F front, a 3-mm-thick Kel-F ring with an inside diameter of 28.6 mm (which formed the liquid cavity), and a 12-mm-thick Kel-F back plate. Copper gauge elements, 5-μm thick and rectangularly shaped (stirrup gauges) with a 50-μm-thick Kapton overlay, were epoxied to the front and back plates so the Kapton was in contact with the liquid. The active length of the gauge was 9 mm. The cell parts were epoxied together and then nylon screws were used to insure that the cell did not come apart under vacuum. Filling the cell with bromoform was accomplished using two small copper tubes epoxied into the back cell plate; these were crimped several times after the filling operation was complete. The cell was mounted to an aluminum backing plate and adjusted (so the tilt was kept below 1 mrad) inside the gun target chamber. An electromagnet, capable of producing a magnetic field of 825 gauss in the cell cavity area, surrounded the target assembly. A Lexan projectile was faced with either Vistal (pressed polycrystalline sapphire) or single crystal sapphire, depending on the impact stress. Aldrich bromoform (Aldrich #24,103-2) was used, as received, with a stated purity of 99+ %; it contained 1% ethanol-C_2H_5OH as a stabilizer.

III. RESULTS AND DISCUSSION

In these experiments, because there were two gauges separated by about 3 mm of bromoform, it was possible to measure both particle velocity in the liquid and the shock velocity of the shock traveling from the first to the second gauge. This made it unnecessary to know the projectile velocity or the Hugoniot of the Kel-F in order to determine a Hugoniot state in the bromoform (assuming no reaction was occurring). Because of this the data are felt to be quite accurate, compared to explosively driven experiments in which impedance matching was required to establish a Hugoniot state (such as was done in the earlier experiments). Data from the five experiments are given in Table 1. These data are plotted, in Fig. 1, in shock-velocity particle-velocity space and compared to the universal liquid Hugoniot for bromoform (using an initial condition sound speed of 0.931 mm/μs)[5] along with Ramsay's data. The waveforms from the four lower pressure experiments show no evidence of reaction. Reaction would be expected to manifest itself in terms of a two-wave structure such as has been previously observed for CS_2[6] and acrylonitrile.[7] Particle velocity waveforms from Shot 745 had noise and strange shapes in the early times of both the front and back gauge records. Because these shapes do not fit with the other higher pressure data and our present understanding of what would be expected if reaction were occurring, we have chosen to treat this experiment as if there were no reaction (until further work is done). This means that up to 9.1 GPa there is no evidence of reaction but there is a possibility something is beginning to happen at 10 GPa.

TABLE 1
NEW HUGONIOT DATA FOR BROMOFORM

Shot Number	Particle Velocity $\frac{mm}{\mu s}$	Shock Velocity $\frac{mm}{\mu s}$	Pressure GPa	Relative Volume $\frac{V}{V_0}$
741	0.534	2.05	3.17	0.740
742	0.680	2.45	4.83	0.723
743	0.840	2.58	6.26	0.674
744	1.067	2.95	9.10	0.638
745	1.138	3.07	10.1	0.629

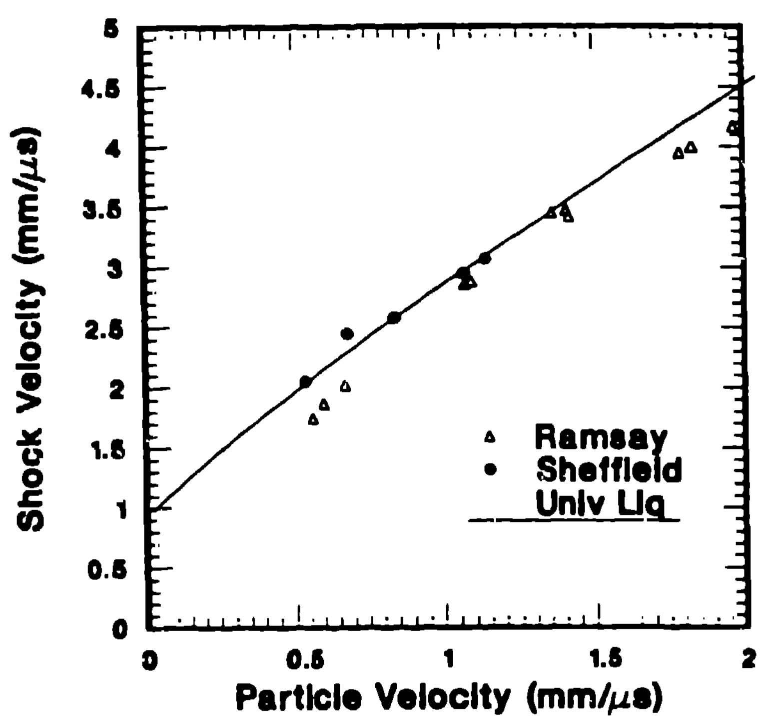

FIG. 1
Hugoniot data for liquid bromoform. Line is universal liquid Hugoniot based on an initial condition sound speed of 0.931 mm/μs. Data are from Ramsay, Ref. 1 and this study.

All five Hugoniot points are quite close to the universal liquid Hugoniot, indicating that this is a very good estimate for the Hugoniot of the material. Ramsay's data deviate from the universal liquid Hugoniot considerably at the low pressures. Whereas we first interpreted this to be possible evidence of low-pressure shock-induced reaction, we now feel there are significant errors in Ramsay's lower pressure data. Since there were no significant deviations from the predicted Hugoniot in our data up to 10 GPa, we believe that the reaction starts at or above this pressure level. It is possible that the bromoform is chemically reacting when it emits in the higher pressure regime where McQueen is using the material. Exactly what the reaction is remains unknown. The data taken at 10 GPa were obtained with the gun shooting at maximum projectile velocity in this type of experiment so it is doubtful that more work will be done until our new 2-stage gun becomes available.

It is perplexing that Ramsay states that bromoform becomes opaque at pressures above 6 GPa with an induction time of 1 to 2 μs since we did not see hydrodynamic evidence of reaction in any of the experiments (with the possible exception of the 10 GPa experiment). This indicates that the reaction which causes the material to become opaque does not have a large enough volume change associated with it to be measurable with particle velocity gauges. The higher pressure data of Ramsay and McQueen lie below the universal liquid Hugoniot, which is evidence that the reaction products are more dense than the bromoform. If this were the case in the lower pressure range, we would expect to be able to measure evidence of this. It is possible that one reaction occurs at low pressures and a different reaction (which emits strongly) occurs at higher pressures. These questions remain to be answered with later work.

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