LA-UR 93-2344

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SUBMITTED TO: Joint AirAPt/APS Conference, June 28 - July 2, 1993
American Physical Society, Colorado Springs, Co

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UNREACTED HUGONIOTS FOR POROUS AND LIQUID EXPLOSIVES[1]

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Numerous authors have measured the Hugoniots of a variety of granular explosives pressed to different densities. Each explosive at each density was typically then treated as a unique material having its own Hugoniot. By combining methods used by Hayes, Sheffield and Mitchell (for describing the Hugoniot of HNS at various densities) with Herrmann's *P*-*α* model, it is only necessary to know some thermodynamic constants or the Hugoniot of the initially solid material and the porous material sound speed to obtain accurate unreacted Hugoniots for the porous explosive. We discuss application of this method to several materials including HMX, PETN, TNT, and Tetryl, as well as HNS. We also show that the "Universal Liquid Hugoniot" can be used to calculate the unreacted Hugoniot for liquid explosives. With this method only the ambient pressure sound speed and density are needed to predict the Hugoniot. Applications presented include nitromethane and liquid TNT.

INTRODUCTION

Equation of state (EOS) information is difficult to obtain on porous explosives as these materials are very sensitive to the shock initiation of detonation. Even at pressures of only a few kbar, shock velocities cannot be obtained from transit time measurements because the explosive is starting to react and accelerate the wave. In wedge tests, wave acceleration is often apparent well before the onset of detonation. Because of the early onset of reaction, measurements of pressure or particle velocity in transmitted waves provide unreliable equation of state information as well.

The best EOS information has been obtained in flying plate experiments using flyers made of well characterized materials whose velocities were accurately measured and when pressure or particle velocity was measured at the flyer/explosive interface. Data from these types of experiments is uncommon, as most researchers have been more interested in the initiation properties, and have hence used wedge experiments.

Because of the difficulties in making reliable EOS measurements on porous explosives it is often useful to construct a Hugoniot using thermodynamic constants obtained from static and hydrostatic measurements, the bulk sound speed of the porous explosive, and then to properly account for the porosity. If hydrostatic data is unavailable, the thermodynamic constants can be obtained by fitting to Hugoniot measurements on single crystals of explosives. These are generally much less sensitive to shock initiation.

POROUS EXPLOSIVE EQUATION OF STATE

The formalism for the equation of state which is used in this paper was developed by Hayes, and was first used to describe the porous explosive, HNS (hexanitrostilbene) by Sheffield, Mitchell and Hayes.[1] It was later amended by Setchell and Taylor[2] to be used in Herrmann's *P*-*α* model.[3] Thus, the ideas are not new, but neither have they seen widespread application.

The method is based on constructing a complete thermodynamic potential function for the fully dense explosive, namely the Helmholtz Free Energy. The specific form which Hayes chose for the Helmholtz Free Energy is[1]

$$F(T,V) = C_V\left[(T - T_0)\left(1 + \frac{\gamma}{V}(V_0 - V)\right) + T\ln\left(\frac{T_0}{T}\right)\right] + \frac{K_{T_0}V_0}{N(N-1)}\left[\left(\frac{V_0}{V}\right)^{N-1} - (N-1)\left(1 - \frac{V}{V_0}\right) - 1\right] \qquad (1)$$

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[1] Work performed under the auspices of the U.S. Department of Energy.

where C_V is the specific heat at constant volume, γ is Gruneisen's parameter, V_0 is the specific volume of the fully dense explosive at zero pressure, K_T is the bulk modulus at constant temperature, and N is a constant. This construction assumes C_V and γ/V are constants, while the bulk modulus is

$$K_T = K_{T_0}\left(\frac{V_0}{V}\right)^N. \tag{2}$$

These constants are readily obtained from static thermodynamic measurements and hydrostatic pressure - volume measurements.

In order to describe the compaction of the porous explosive in the framework of Hermann's P-α model,[3] Eq. (1) must be recast in the form $P(E,V)$ or $E(P,V)$. Setchell and Taylor[2] have shown that

$$\begin{aligned} E(P,V) = {} & \frac{P}{\gamma/V} - \frac{\gamma}{V} C_V T_0 (V_0 - V) \\ & - \frac{K_{T_0}}{N\gamma/V}\left[\left(\frac{V_0}{V}\right)^N - 1\right] \\ & + \frac{K_{T_0} V_0}{N(N-1)}\left[\left(\frac{V}{V_0}\right)^{1-N} \right. \\ & \left. - (N-1)\left(1 - \frac{V}{V_0}\right) - 1\right] \end{aligned} \tag{3}$$

Eq. (3) describes the behavior of the fully dense explosive. While Eq. (1) is a complete thermodynamic EOS, Eq. (3) is not.

Herrmann's theory,[3] as modified by Carroll and Holt[4] states that in a porous material, Eq. (3) describes the solid portion. The distension of the porous material is

$$\alpha = V/V_s, \tag{4}$$

where V is the specific volume of the porous explosive at a given pressure and energy, and V_s is the specific volume of the solid explosive at the same pressure and energy. If in the porous material the average pressure is P and the average specific volume V, then the pressure in the solid portion is $P\alpha$ and the specific volume of the solid portion is V/α. Equation (3) can thus be used to describe the energy at average pressure P and volume V in the porous explosive if modified to describe only the solid portion:

$$E = E(P\alpha, V/\alpha). \tag{5}$$

Eqs. (3), (4), and (5) are now augmented by defining α as a function of the shock pressure P. A simple form for $\alpha(P)$, such as the following one suggested by Herman,[3] is sufficient.

$$\begin{aligned} \alpha &= 1 + (\alpha_0 - 1)(1 - P/P_s)^2 \cdots (P \le P_s) \\ \alpha &= 1 \qquad\qquad\qquad\qquad\quad \cdots (P \ge P_s) \end{aligned} \tag{6}$$

In Eq. (6), α_0 is the original distension, $\alpha_0 = V_{00}/V_0$, and V_{00} is the initial specific volume of the porous explosive. P_s is the pressure at which the material crushes to solid density. Herman has also shown how P_s can be determined from measurements of the sound speed in the porous material.[3] Different values of P_s must be used for different initial porosities.

Given P and $\alpha(P)$, Eq. (5) is then itteratively solved for V such that the energy is the same as that given by the Hugoniot jump condition $E = P/2(V_{00} - V)$. Given the quality of much of the experimental EOS data, further sophistication in an EOS is unwarranted.

POROUS EXPLOSIVE DATA

Table 1. lists the thermodynamic constants for the explosives HMX, HNS, PETN, TNT, and Tetryl. Brief comments about where these constants were obtained, and how well they fit the data follow.

K_{T_0} and N for PETN were obtained by fitting the hydrostat obtained by Olinger and Cady.[5]

Table 1. Thermodynamic constants by explosive.

Explosive	ρ_0 g/cm³	K_{T_0} Mbar	N	γ/V g/cm³	C_V cm²/µs²/K
HMX	1.90	0.129	10.3	2.09	$1.05(10^{-5})$
HNS	1.74	0.146	3.5	2.82	$0.89(10^{-5})$
PETN	1.77	0.110	7.1	2.04	$1.00(10^{-5})$
TNT	1.65	0.082	7.3	1.51	$1.08(10^{-5})$
Tetryl	1.73	0.072	10.2	1.56	$1.08(10^{-5})$

We have also used the values of γ , and C_V contained in Ref. 5. Using these values and the crystal density of 1.775 g/cm^3 reproduces Shock Hugoniot measurements at 1.775, 1.75, and 1.71 g/cm^3 [6] fairly well, as shown in Figure 1 and 2. Using the P-α formalism with values of P_s = 15 and 3.5 kbar respectively, also reproduces the 1.60 and 1.40 g/cm^3 data shown, which were selected from the reports of Wackerle and coworkers.[7,8] These data were obtained using x-cut quartz and manganin pressure gauges. As porous explosive Hugoniot data go, these are excellent. As is typical there is more scatter in the P-V than the U_s - u_p plane.

K_{T_0} and N for HMX were obtained from fits to the hydrostatic pressure - volume data of Olinger, Roof and Cady.[9] γ and C_V are as reported in Ref. 9, as well. These constants, reproduce the 1.891 g/cc HMX Hugoniot[10] quite well up to pressures of about 100 kbar. Above 100 kbar, neither the 1.891 nor the single crystal HMX Hugoniot[11] is reproduced very well, possibly because of a phase transition in the HMX at a pressure near 100 kbar.[12] These constants with P_s set equal to 2.5 kbar nicely reproduce our Hugoniot measurements on 1.24 g/cc HMX.

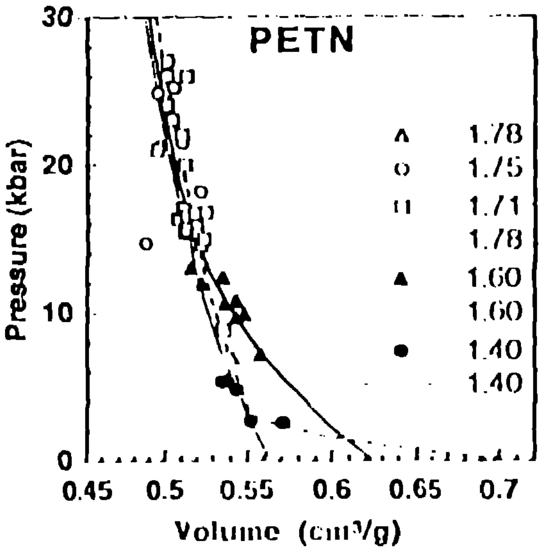

Figure 1. Hugoniots for PETN in the P-V plane. The initial density is noted beside each symbol. Smooth curves were calculated using the initial density noted beside the curve symbol. P_s was 3.5 kbar for the 1.40 g/cm^3 PETN and 15 kbar for the 1.60 g/cm^3 PETN.

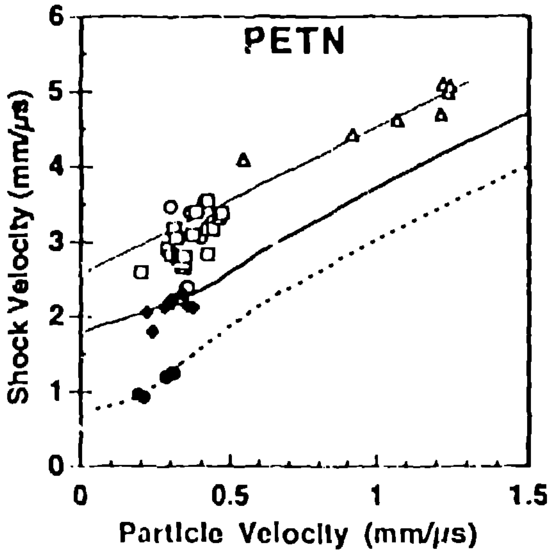

Figure 2. Hugoniots for PETN in the U_s - u_p plane. Symbols represent the same data sets as those in Figure 1. Solid curves are calculations. The data shown in this figure cover a wider pressure range than those in Figure 1.

The constants we show for the γ and C_V of HNS were obtained from Setchell and Taylor.[2] While Setchell and Taylor[2] also claimed to have improved estimates for K_{T_0} and N, we found that they do not reproduce the data of Ref. 1. as well as the original constants reported by Sheffield et. al.[1] Hence we list K_{T_0} and N from Ref. 1.

For TNT, γ and C_V, were obtained from values for the volume coefficient of thermal expansion and C_p [13] combined with values of K_{T_0} and N obtained from fitting to single crystal TNT Hugoniot data.[14] Relations among the thermodynamic constants were used with an iterative procedure to arrive at the final values.

For Tetryl, good values of the thermodynamic constants were not available in the literature. Because of its chemical similarity with TNT, we set γ and C_V equal to the TNT values. K_{T_0} and N were then obtained by least squares fitting to the 1.70 g/cc (96.3 % of crystal density) Hugoniot data of Lindstrom.[15] Lindstrom's data for lower densities of tetryl were obtained from wedge experiments and have significant scatter (in the P-V plane).[15]

LIQUID EXPLOSIVES EQUATION OF STATE

Liquid explosives and many other liquids have equations of state, which for the purpose of predicting shock states are well defined by the "Universal Liquid Hugoniot" first proposed by Woolfolk, Cowperthwaite, and Shaw.[16]

$$\frac{U_s}{C_0} = 1.37 - 0.37e^{-2u_p/C_0} + 1.62\frac{u_p}{C_0} \qquad (7)$$

In Eq. (7), U_s and u_p are, as usual, the shock and particle velocity, while C_0 is the sound velocity at zero pressure. Figure 3 shows measured shock and particle velocities for the explosives liquid TNT[17] and nitromethane.[18-20] The solid curves were calculated using Eq. (7) and C_0 = 1.55 mm/μs for liquid TNT[21] and C_0 = 1.30 mm/μs for nitromethane. Reproduction of the experimental data is excellent. Unfortunately, virtually no Hugoniot information exists for other liquid explosives. However, we have found that Eq. (7) works well for every other liquid we have tried.

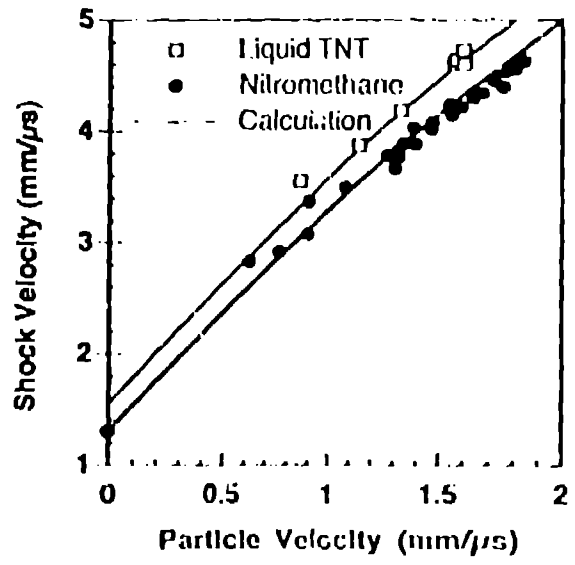

Figure 3. Hugoniots for liquid TNT and nitromethane in the $U_s - u_p$ plane. Experimental data is from Garn[17] for liquid TNT and from Refs. (18-21) for nitromethane. Solid lines were calculated from Eq (7).

ACKNOWLEDGMENTS

We would like to thank Ronald Rabie for useful discussions concerning the P-α model, and Jerry Dick and Jerry Wackerle for discussions concerning general properties of porous explosives.

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