LA-UR- 98-3365

CONF-980803--

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Submitted to: 11TH DETONATION SYMPOSIUM, SNOWMASS, CO AUG 30 - SEP 4, 1998

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A STUDY OF THE OVERDRIVEN BEHAVIORS OF PBX 9501 AND PBX 9502

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We present the Hugoniot pressure and sound speed data in the overdriven regime for both PBX 9501 and PBX 9502. The overdriven release experiments are also given. The failure of the standard Jones-Wilkins-Lee equation of state in modeling both the Hugoniot data and the overdriven release experiments for both high explosives are identified and remedy is made by including additional terms to steepen the slope of the Hugoniot in the high pressure regime. However, an anomaly presented itself as a kink in the release wave of PBX 9502 is observed. A careful examination of the Hugoniot data indicates similar behavior. We suggest a possible explanation for this peculiarity to the phase transition of carbon in the products.

INTRODUCTION

In an ideal hydrodynamic process behind the detonation front of high explosives (HE), the detonation products follow a monotonically isentropic expansion pathway beginning at the Chapman-Jouguet (CJ) state. The effort to develop an equation of state (EOS) representing the products' behavior is essentially the determination of the principal isentrope below the CJ point. The EOS is required to be valid only in a narrow band enclosing the isentrope. Closely resembling this picture is the cylinder test that forms the basis of most EOS calibration, including the well-established Jones-Wilkins-Lee (JWL) form.[1] However, even in such a simple case, the reflection of the detonation wave from the cylinder wall, usually copper, perturbs the expansion from the ideal pathway by some degree of recompression in the interior of HE. In most engineering systems, stronger reflection can occur, depending on the configuration and the boundary materials. The reflection can result in a hydrodynamic state that deviates substantially from the principal isentrope, and the pressure can be higher than the CJ value. Also collision of detonation waves originated from various sources can produce a very high pressure effect. The hydrodynamic regime as mentioned is not replicated by the simple cylinder test, and the knowledge needed for the EOS is beyond the principal isentrope. That is one of the reasons why we cannot use the same JWL EOS based on the cylinder test only in other geometries. The other major issue is the reaction zone effect in which the reaction has not completed at the detonation front in the time and space scales compatible to the hydrodynamic event. Therefore, the kinetics aspect, namely, the change of compositions that follows a finite rate, must be addressed along with the already complicated products EOS. In this work, we report results of new data of shock pressure and sound speed above the CJ State for two fundamentally different types of HE, and the development of a modified JWL EOS to correct the deficiency found in the standard formulation. Finally, we use the new form of EOS to simulate a new class of hydrodynamic experiments that provides an overdriven condition first and then the flow expands to lower pressure level isentropically. The entire process is more relevant to the flow regimes found in many engineering systems while it still remains simple enough for easy diagnostics. From this investigation, we can find out how well the new EOS performs in a more realistic situation and, perhaps most important of all, we can find any surprises. It turns out for one HE the latter is to be the case.

OVERDRIVEN HUGONIOT PRESSURES AND SOUND SPEEDS

The overdriven regime is loosely defined as the hydrodynamic condition above the CJ pressure. We do not present the techniques of obtaining overdriven Hugoniot and sound speed experiments here since they have been documented elsewhere.[2] This paper concentrates on the validity of the standard JWL EOS in matching the overdriven data. First we review briefly the standard JWL EOS formulation. Instead of writing a global EOS format, we prefer the formulation using the principal isentrope as the reference:

$$p_i = A\ e^{-R_1 v} + B\ e^{-R_2 v} + C\ v^{-(1+\omega)} \tag{1}$$

$$\varepsilon_i = \frac{A}{R_1}\ e^{-R_1 v} + \frac{B}{R_2}\ e^{-R_2 v} + \frac{C}{\omega}\ v^{-\omega} \tag{2}$$

p, ε, and v are the pressure, specific internal energy and relative volume. The subscript i indicates the quantity on the principal isentrope. The rest are material property constants. The complete equation of state is expressed as follows.

$$p = p_i + \frac{\Gamma}{v}\ (\varepsilon - \varepsilon_i) \tag{3}$$

We deliberately select a different symbol for the Gruneisen gamma Γ here from the one on the isentrope, ω, the commonly appears in standard formulation. The reason will be made clear later.

Once the general form of the EOS is known, the shock Hugoniot can be found

$$p_H = [p_i + (\varepsilon_0 - \varepsilon_i)] / \left[1 - \frac{\Gamma}{2v}\ (1 - v)\right] \tag{4}$$

where

$$\varepsilon_0 = \varepsilon_{cj} - \frac{1}{2}\, p_{cj}\ (1 - v) \tag{5}$$

The sound speed on the isentrope is

$$c_i^2 = -\frac{v^2}{\rho_0}\frac{dp_i}{dv} \tag{6}$$

and the one on the Hugoniot

$$c_H^2 = c_i^2 + \frac{v}{\rho_0}(p_H - p_i)\left(\Gamma + 1 - \frac{v}{\Gamma}\frac{d\Gamma}{dv}\right) \tag{7}$$

Subscripts H and cj represent the one on the Hugoniot and at the CJ point. A nonconstant Γ, a function of volume, is anticipated, as we shall see in later development.

Traditionally, the shock pressure on the Hugoniot is the only information obtained from the basic shock experiments for HE. The sound speed information, although widely recorded for inerts, is usually not available. Only recently, effort has been made toward that goal but only for a limited situation. The significance of the sound speed in an HE system can be seen as follows. Since signal travels at sound speed, any erroneous signal traveling time leads to a false information on the actual HE boundary location and the boundary material, for example. So in an engineering system, the wrong timing of various signals from the HE boundaries effectively alters the geometry. A correct EOS must address this issue as well, since the sound speed, a derivative quantity, is much more difficult to obtain experimentally than the shock intensity.

We have previously examined the adequacy of the standard JWL EOS in matching the overdriven Hugoniot experiments for PBX 9501 (95% HMX, 2.5% Estane, 2.5% BDNPA/BDNPF).[3] The JWL EOS underestimates the shock pressure by about 30 *kbar* in the 600 *kbar* range. The error is about 5 percent. However, even at a moderate pressure range slightly above CJ estimated at 360 *kbar*, the calculated sound speed is noticeable below the experimental value. The deficiency is about 0.8 *mm/μs*, roughly 10 percent of the experimental value at 600 *kbar*. The discrepancy worsens as the pressure goes up higher. Here we see a good example that the JWL EOS performs adequately in predicting the shock pressure in the moderate overdriven regime but poorly in calculating the sound speed even in a slightly overdriven condition. The reason for the failure of the JWL EOS in matching experimental results in many engineering systems is more likely the failure of timing due to the sound speed error, rather than the weaker force field caused by a slightly lower pressure. Unfortunately, these two aspects are not easy to separate in many integrated experiments without careful instrumentation.

MODIFIED JWL EQUATION OF STATE

This report is not the first time the deficiency of the JWL EOS in dealing with the overdriven hydrodynamic regime has been reported.[4,5] Previous works on the subjects have been investigated and some degrees of improvement has been also been suggested mainly by adding more exponential terms.[6] In this paper, we propose a different formulation from previous ones. The main objective is to preserve the original set of parameters developed from other reliable means such as the calibration from the cylinder test below CJ. Of course, we have to assume at least in principle that the low-pressure portion of the EOS is acceptable. Again, the experimental nature of this work does not assure uniqueness in final result.

Since the deficiency of the JWL EOS is found mainly in the overdriven regime, we propose to steepen the slope of the Hugoniot to match the Hugoniot data better and by doing so we hope to increase the sound speed as well. The idea of the formulation is similar to the concept of compressibility in perfect gas in the high-pressure region. We apply corrections in the form of multipliers to the highest exponential terms only

$$p_i = [1 + F_p(v)]\, A\, e^{-R_1 v} + B\, e^{-R_2 v} + C\, v^{-(1+\omega)} \quad (8)$$

$$\varepsilon_i = [1 + F_\varepsilon(v)] \frac{A}{R_1} e^{-R_1 v} + \frac{B}{R_2} e^{-R_2 v} + \frac{C}{\omega} v^{-\omega} \quad (9)$$

The corrections are in simple polynomial form of the difference between the current volume and the CJ volume.

$$F_p(v) = A_0 (v_{cj} - v)^2 + B_0 (v_{cj} - v)^3 \quad (10)$$

$$F_\varepsilon(v) = \left[A_0 - \frac{3B_0}{R_1}\right]\left\{\frac{2}{R_1^2}\left[1 - e^{-R_1(v_{cj} - v)}\right] - \frac{2}{R_1}(v_{cj} - v) + (v_{cj} - v)^2\right\} + B_0 (v_{cj} - v)^3 \quad (11)$$

The correction is applied only when the volume is less than the CJ volume, namely, stronger compression than the CJ state. There are only two new parameters, A_0 and B_0, appearing in the principal isentrope for both the pressure and the internal energy. The calibration requires the Hugoniot data for a fixed Γ. However, we find that any reasonable choice of Γ is acceptable, so additional constraint must be imposed. It turns out that the sound speed provides such a condition. Figure 1 shows the results of sound speed calculation for PBX 9501 using three different Gruneisen Γ, 0.20, 0.38, and 0.80. In this case the nominal value 0.38 based on the cylinder test is found to be the most proper one to match the sound speed data. A different approach but still using the overdriven portion of the sound speed information only concludes a slightly different value at 0.45.[2] In addition, the sound speed data can be used to determine the CJ pressure. Figure 2 presents the results of 370, 360 and 355 *kbar*, and we conclude that the CJ pressure should be about 360 *kbar*. After we make the choice on CJ pressure and Γ, Figure 3 shows the overdriven Hugoniot data of PBX 9501 and the Hugoniots before and after the conventional JWL EOS is modified using the normal value for Γ. And for the

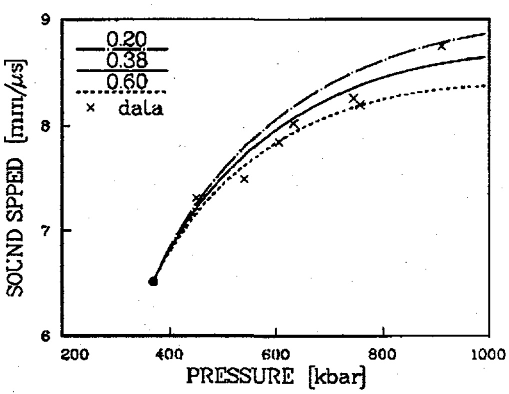

FIGURE 1. PBX 9501: EFFECT OF GRUNEISEN GAMMA ON THE SOUND SPEED.

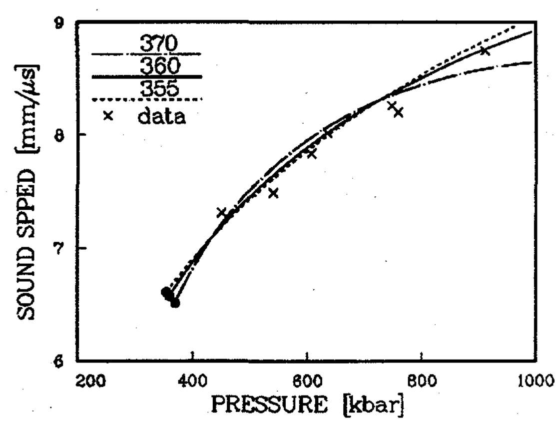

FIGURE 2. PBX 9501: EFFECT OF CJ PRESSURE ON THE SOUND SPEED.

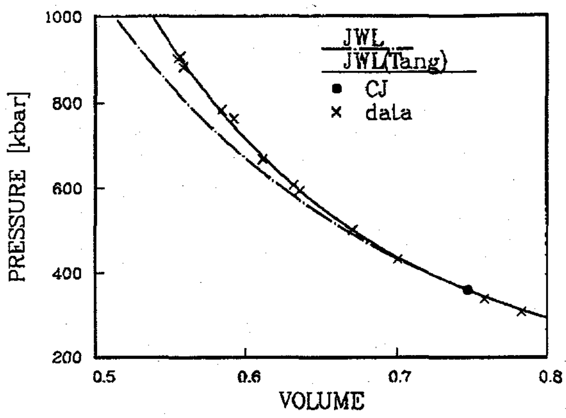

FIGURE 3. PBX 9501 OVERDRIVEN HUGONIOT.

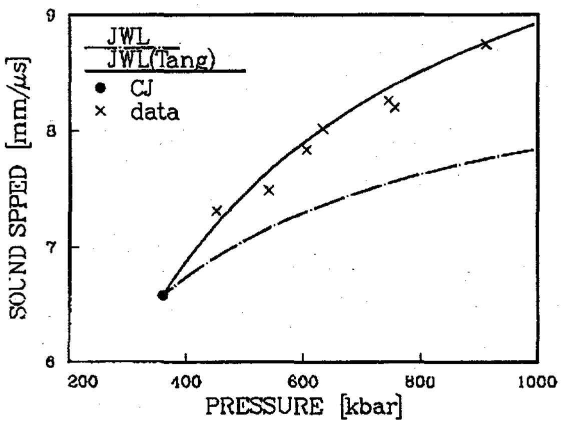

FIGURE 4. PBX 9501 OVERDRIVEN SOUND SPEED.

sound speed, without the correction terms, the nominal JWL EOS fails miserably in this range, as seen in Fig. 4. We must be cautioned at this stage not to rely so much on the sound speed information due to a fairly large error bar in the experimental measurement.

Another HE we have great interest in is PBX 9502 (95% TATB, 5% Kel-F). It is known for its nonideal behavior due to a large amount of carbon in the products, and it has a significant reaction zone size, about 2 *mm*, in steady detonation. The poor performance of the standard JWL EOS in matching the Hugoniot pressure at moderate pressure level above the CJ condition, estimated at 285 *kbar*, is quite obvious. Even at 400 *kbar*, the standard JWL EOS is short by close to 5 percent in shock pressure. In view of the success with PBX 9501, without hesitation we proceed to apply the modified JWL EOS formulation. Figure 5 shows the Hugoniot pressure

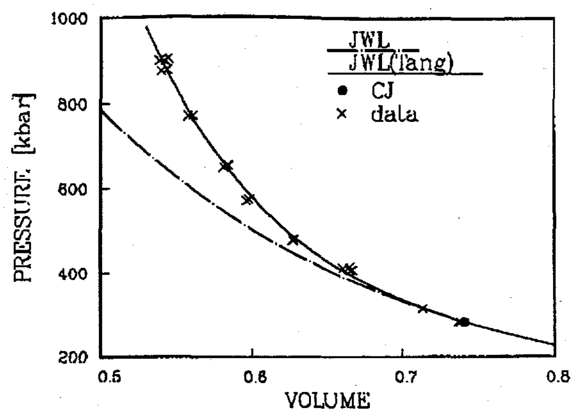

FIGURE 5. PBX 9502 OVERDRIVEN HUGONIOT

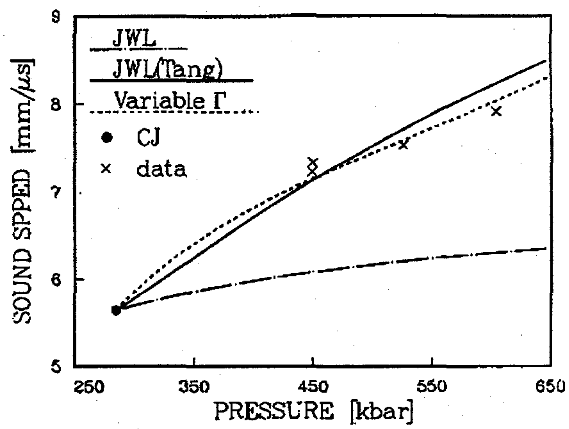

FIGURE 6. PBX 9502 OVERDRIVEN SOUND SPEED

results from experiment and the two JWL EOS calculations; and the improvement of the modified JWL EOS formulation is evident. Along the same direction, the sound speed data are collected and presented in Fig. 6. With a limited amount of data available due to the reaction zone effect that complicates the experiment, we find that a constant Γ is not sufficient to reproduce the experimental result. Instead, a variable Γ is required and increases as volume decreasing.

OVERDRIVEN RELEASE EXPERIMENTS

Recently we developed a new type of experiment to produce the overdriven behavior of the HE products in an environment compatible to what is expected in a real engineering system and then to examine the adequacy of the standard JWL EOS based on the cylinder test in the high-pressure regime. An explosive-launched flyer not only initiates but also maintains the pressure of the target HE at the level above CJ. The thickness of the flyer keeps the pressure constant over a period of time in the interior of the HE until the rarefaction wave from the back of the flyer reaches the same location and the pressure starts to decrease. The expansion is quite similar to the one in a normal detonation except that the initial state is on the principal Hugoniot above CJ. The hydrodynamic state is monitored by measuring the interface velocity between the HE and a transparent window such as LiF. There are two major quantities to look for. One is the constant velocity portion, which represents the overdriven condition. The other is the delay time between the arrival of the detonation wave represented by the initial velocity jump and the onset of the velocity decay as the rarefaction wave reaches that interface. This delay time is a good measure of the average sound speed in the overdriven regime. However, it should be cautioned that the sound speed is not constant all the way due to the reaction zone effect. Closer to the front, the sound speed is higher but should approach to a steady value before the rarefaction moves in.

The simulation of the overdriven release experiments for the PBX 9501 using the standard JWL and the modified JWL are compared first. A piece of 13.018-*mm* long PBX 9501 is initiated by an explosive-launched Al flyer of 4.711-*mm* thick and velocity at 5.414 *mm*/μ*s*. A detonation wave of 520 *kbar* is produced inside the PBX 9501. At the end of the HE charge, a transparent window of LiF window is employed so that the interface velocity can be measured using VISAR. There is no major difference between the regular JWL and the modified one in the 500-*kbar* range as confirmed by the Hugoniot data, see Fig. 7. But the major improvement is seen in a shorter time interval between the arrival of the detonation wave and the lead rarefaction wave as the result of the higher calculated sound speed in the new EOS formulation than the old one. We expect to see more significant effect from the sound speed for longer HE charge and certainly at higher pressure level.

We repeated similar experiments on PBX 9502. The first one was with flyer velocity 5.27 *mm*/μ*s* and flyer thickness 4.70 *mm* and gave an initial shock of 480 *kbar*. The improvement is seen in the drastic shortening of time interval between the detonation wave arrival and the rarefaction wave arrival even for a short HE charge length of 12.9 *mm*. Also a significant improvement is observed in the shock

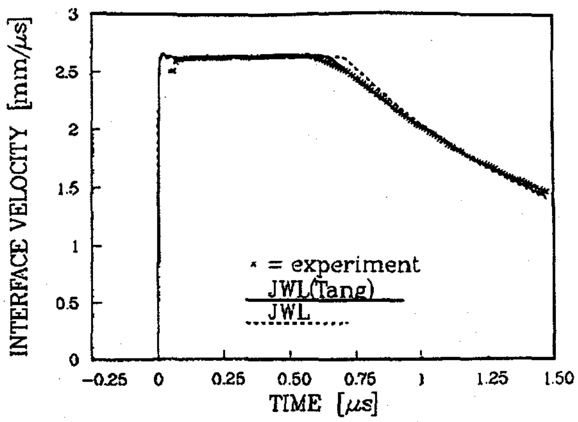

FIGURE 7. PBX 9501 OVERDRIVEN RELEASE WAVE WITH INITIAL PRESSURE AT 520 KBAR.

pressure level as indicated by a higher interface velocity in the flat portion of the wave using the improved JWL EOS, given in Fig. 8.

Optimism dwindles somewhat when we compare the new JWL EOS result to the experimental data down the pressure level. The calculation matches the experiments quite well until the velocity or the pressure has dropped to a certain value. The experiments indicate a faster decay rate than the calculated one below that point as the slope of the release wave changes rather suddenly. This changeover that appears as a kink occurs around 400 *kbar* and is quite reproducible as we conduct another experiment with a similar geometry and condition. The same phenomenon is seen. Interestingly enough, the new EOS calculation helps to locate the abnormality in the experiment not easily identifiable by the experimental record alone. We then conduct more experiments at lower shock and shorter duration using a flyer with thickness of 3.20 *mm* and velocity of 4.70 *mm*/μ*s*. The condition produces a shock level about 415 *kbar*; the results are shown in Fig. 9. First we see the reaction zone effect as indicated by the spiky peak which is consistent with previous reaction zone experiments in the unsupported detonation.[7,8] But then we observe the same kinky transition behavior also in the 400 *kbar* range. This peculiar behavior is not seen in PBX 9501 over the same pressure range compatible to those on PBX 9502. We even perform experiments by replacing the HE with Al and to look at the Al-LiF interface velocity history as the system is impacted by an Al flyer. The release wave behaves, as it should without wriggle. So we can conclude with confidence that the kink is an indication of the products characteristics peculiar to PBX 9502 in the 400 *kbar* range. Here we are cautioned that the reference to the value of 400 *kbar* is

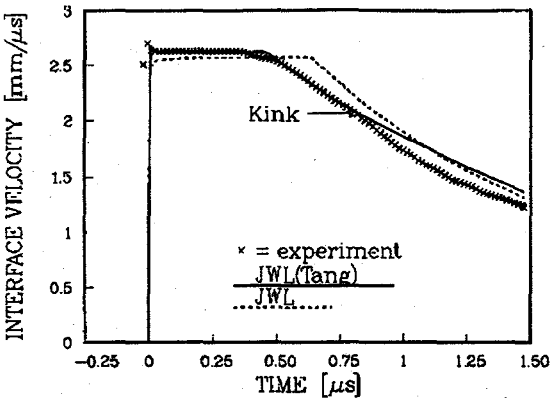

FIGURE 8. PBX 9502 OVERDRIVEN RELEASE WAVE WITH INITIAL PRESSURE AT 480 KBAR.

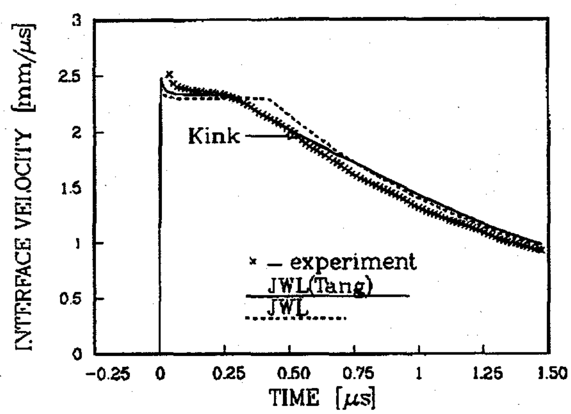

FIGURE 9. PBX 9502 OVERDRIVEN RELEASE WAVE WITH INITIAL PRESSURE AT 410 KBAR.

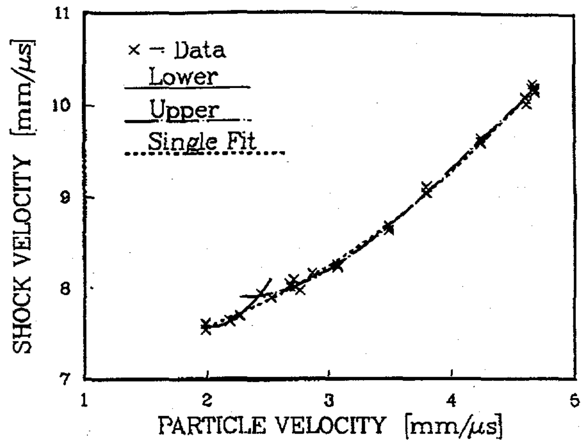

FIGURE 10. PBX 9502 OVERDRIVEN HUGONIOT IN SHOCK VS PARTICLE VELOCITIES.

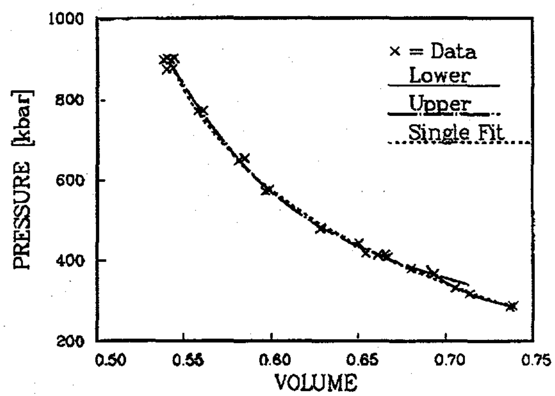

FIGURE 11. PBX 9502 OVERDRIVEN HUGONIOT IN PRESSURE VS VOLUME.

only a matter of convenience to indicate an unusual event taking place in the range. There is no implication whatsoever about the exact condition for the event to occur.

After the discovery, we gather more Hugoniot data to see what is going on in the pressure range around 400 *kbar*. The result is shown in Fig. 10 for shock velocity versus particle velocity with more data in the range. A careful examination of the Hugoniot data reveals that it does not follow a smooth curve. In fact, the data can best be fitted with two segments. The change of the slopes of the two segments is quite clear. The intersection of the two segments turns out to be near but below 400 *kbar* as given in the pressure versus volume plot, Fig. 11, although it is not so clear as in the shock velocity vs particle velocity plot. Of course the precise intersection of the two branches depends on the amount and the quality of the data available in that neighborhood. So the kink appearing in the overdriven release wave is not a special case and is definitely consistent with the Hugoniot data. There is something unusual happening at that pressure level in PBX 9502.

Since the kink is not seen in PBX 9501 but only in PBX 9502, we try to connect this peculiar behavior to the abundance of carbon found in the products of PBX 9502. We have identified in the past that the extended reaction zone in PBX 9502 is caused by the slow coagulation of carbon which has been incorporated in a unified reaction kinetics model.[9,10,11] In the high pressure regime, we wonder if carbon could also be the culprit in contributing to such a kinky behavior in PBX 9502 as seen in the overdriven release wave and also in the Hugoniot. Since there are no overdriven release experiments available for other

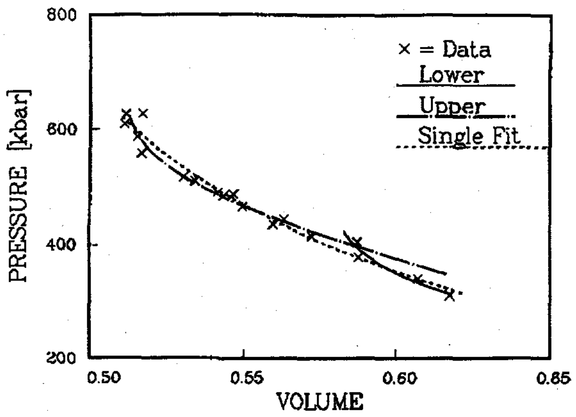

FIGURE 12. TNT OVERDRIVEN HUGONIOT IN PRESSURE VS VOLUME.

carbon-rich HEs, we are inclined to search for the overdriven Hugoniot information. We find data available for Trinitrotoluene (TNT).[4] Although the original paper did not report such an unusual behavior, after a careful analysis, we believe it does exhibit the type of changeover similar to PBX 9502, and in fact even more so. Figure 12 clearly shows that the two segment fits are much better than the single curve fit. And the pressure at the intersection also falls around 400 *kbar*. If we trust the data we are getting, then the Hugoniots of PBX 9502 and TNT indeed exhibit the type of characteristics associated commonly with phase transition. So we are inclined to speculate the likelihood of phase transition linked to carbon, a new discovery in determining the behavior of the HE detonation products. Furthermore, the overdriven condition could result in the melting of carbon, and the transition could involve the liquid to solid phase change as the products expand and cool off. We need more experimental work to support this postulation.

CONCLUSIONS

We construct an extension of JWL EOS and fit the new form using the overdriven Hugoniot and sound speed data. Then we check the adequacy of the new EOS in simulating the overdriven release experiments. So far it works very well for PBX 9501. However, the new EOS is not sufficient to model the more complex behavior of PBX 9502 we discover in this investigation. More work is ahead of us in trying to understand the behavior of HE detonation products, experimentally and theoretically. For the hydrodynamic simulation, the construction of an equation of state reflecting such a complexity is essential if it is for practical purpose. We need more hydrodynamic data including sound speed information. The sound speed perhaps is the best indicator of any irregularity in the overdriven behavior. The difficulty of obtaining good sound speed data using our current technique is the effect of the reaction zone when the condition is close to the CJ state. New experimental techniques must be developed to look for the possibility of carbon phase transition in HE products or other possible mechanisms with similar behaviors.

ACKNOWLEDGMENTS

The author wishes to thank W. Anderson, J. Fritz, R. Hixson and J. Vorthman for the data. The work is supported by the United States Department of Energy under contract W-7405-ENG-36.

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