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TITLE: A STUDY OF THE IMPACT OF REACTION RATES ON EQUATION OF STATE

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A STUDY OF THE IMPACT OF REACTION RATES ON EQUATION OF STATE

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We present experimental evidence on high explosives pushing metal plates that shows the tendency of the equation of state behaving more energetically in the low-pressure region. We believe the deficiency in predicting low- and also high-pressure performance has a theoretical origin, and it is related to reaction rate. The reaction process with a slow stage generates a higher effective Chapman-Jouguet pressure. The development of an equation of state based on this information, using the classical detonation theory but without the consideration of the slow component in reaction, would produce a more energetic condition in the low-pressure region and a less powerful one in high pressure. We demonstrate how a polytropic gas equation of state is constructed and the properties it possesses as mentioned. Finally, we show how the effective Chapman-Jouguet condition comes about and define the property of a pseudo Hugoniot associated with it.

1. INTRODUCTION

Extending the range of applications beyond the domain of calibration is a way of life in engineering and science. In high explosives (HE), we do not usually generate very high pressure in simple experiments, and in other extremes, very low pressure is not easily attainable either. Nevertheless, extreme conditions of high and low pressure are often encountered in many applications. To these situations, data developed for the moderate ranges are used.

In previous work through the simulation of plate push experiments,[1] we described how the slow process stage in the reaction can affect the Chapman-Jouguet (CJ) state. It brings about a higher effective pressure than the normal value based on simple detonation theory. Only in passing did we mention the deficiency of the EOS in the low-pressure region without giving any reason. The product equation of state used is Becker-Kistiakowsky-Wilson (BKW), but the source of the defect is not unique to this particular EOS.

Figure 1 shows the free surface velocity histories of both the experimental and computational results for a 50-mm thick PBX 9502 (95% TATB, 5% kel-F 800) pushing aluminum (Al) and tantalum (Ta) plates of 0.5-mm thickness. Without the inclusion of a slow process, the prediction would fall short of the experiment in the initial velocity jump, indicating a lower CJ pressure condition.[1] With the proper reaction rate, good match is obtained throughout for Ta; but for Al, the simulation predicts a higher velocity after a couple of reverberations. Certainly we could blame the inadequacy of the EOS for Al, but we do have high confidence in the EOS used because it is obtained from a wide range of experiments. So the fault must be the HE EOS. To see why this deficiency occurs only when Al is used, we examine

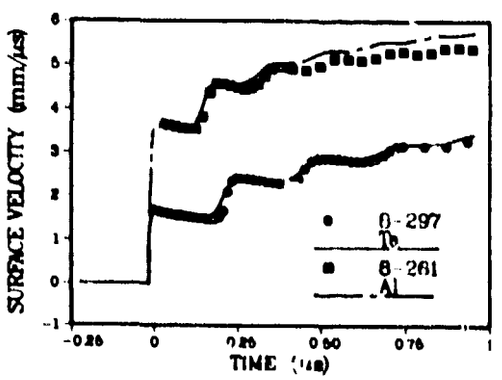

FIGURE 1
50-mm PBX 9502 pushing aluminum and tantalum plates.

the pressure time histories at the explosive-metal interface as seen in Fig. 2. Because of the high impedance Ta, the pressure is still above 10 GPa around 1 μsec; but the condition is not so for the low impedance Al. Pressure in the Al case already drops below 5 GPa after the arrival of the second release wave. Similar behavior is seen when a 13-mm PBX 9502 is used.[1]

Additional evidence is seen in interface velocity experiments, especially for short-duration impact and low-impedance window materials.[2] At this point we begin to suspect the inadequacy of the HE EOS at low pressure, but the question remains, why is it more energetic? Is it merely a condition of defect in numerical extrapolation, or does it have any intrinsic physical significance? This work is to answer the question, if not to offer final solution.

2. POLYTROPIC GAS EQUATION OF STATE

To afford our analytical investigation, we have to select the simplest equation of state, such as polytropic gas, although we recognize the inadequacy of this particular EOS for condensed phase explosives. However, the most common use of EOS, Jones-Wilkins-Lee (JWL), does have an asymptotic behavior of the polytropic gas at low pressure. The EOS is[3]

$$pv = (\gamma - 1)(e + \lambda q); \tag{1}$$

and the CJ property

$$p_{cj} = D^2/v_o(\gamma + 1), \tag{2}$$

and

$$D^2 = 2(\gamma^2 - 1)q. \tag{3}$$

The initial pressure is neglected in the formulation; p, v, and e represent pressure, specific volume, and specific internal energy; while q is the heat release, γ the polytropic coefficient, and λ the reaction fraction. D is the detonation velocity; subscript cj refers to CJ state, and subscript o to initial value. It is a simple matter to construct a polytropic gas EOS if CJ pressure and detonation velocity are known. Choosing dimensionless unit, $v_o = 1$, and $D^2 = 80$, we have for two different CJ pressures,

$$p_{cj} = 32., \quad \gamma = 1.50;$$

$$p_{cj} = 36., \quad \gamma = 1.22.$$

The two Hugoniots along with a common Rayleigh line are shown in Fig. 3. We see quite clearly that the Hugoniot with higher CJ pressure is more energetic than the other in the low-pressure region but less energetic in the high-pressure region, provided they have the same detonation velocity and, therefore, the same Rayleigh line. As we have already noticed in Eq. (1), the form of EOS should show dependence on the reaction fraction λ, so in general,

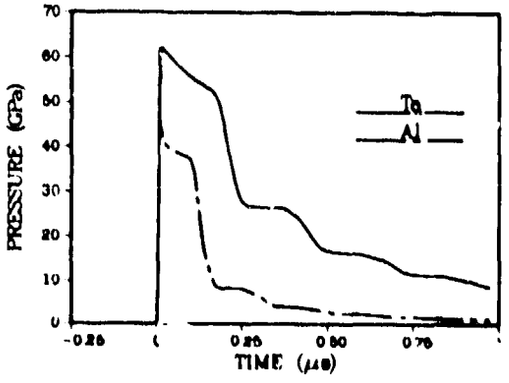

FIGURE 2
Pressure histories at interfaces between HE and metal plates.

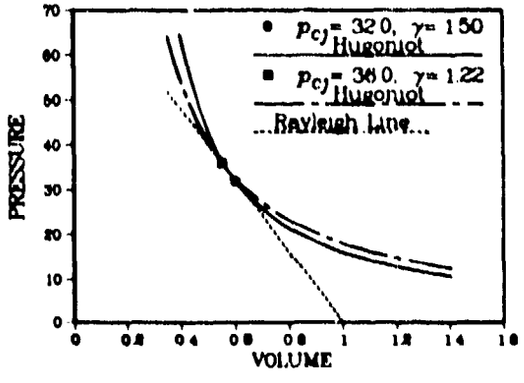

FIGURE 3
Construction of a Hugoniot based on CJ state for a polytropic gas.

$$p = p(e, v, \lambda) . \qquad (4)$$

The appearance of λ is critical in expressing the EOS of energetic materials. Only in chemical equilibrium, which assumes infinite rates, can explicit dependence of p on λ be removed.

3. EFFECTIVE CHAPMAN-JOUGUET STATE AND PSEUDO HUGONIOT

All equations of state require some sort of normalization or parameter adjustment to fit experimental data. In the previous section we have demonstrated such an approach for known detonation velocity and CJ pressure. However, CJ pressure is not a directly measurable quantity and is inferred only from experiments such as plate push. As we have demonstrated in the simulation of plate push experiments, a slow component must be added to the reaction process in order to fit the experimental data better.[1] The consequence of this reaction step is to make the CJ pressure "appear" higher than the normal CJ pressure, for the reason given later. Since we can only measure the "effective" CJ pressure, not the normal one, any construction of EOS using the effective value based on classical detonation theory will lead to wrong result, regardless of the types of EOS.

We determine through interface velocity experiments, as well as plate push experiments, that the detonation of condensed high explosives consists of a fast reaction stage followed by a slow reaction stage. Figure 4 shows such a reaction history for PBX 9502. The first 85 percent is burned in less than 20 ns, but it takes more than 380 ns to reach the level of 99.9 percent. The question remains why the presence of a slow process would make explosives behave more energetically in the low-pressure region as implied through plate push experiments. Referring to Fig. 5, we proceed with the argument. In the $p - v$ plane, first there are two frozen Hugoniots labeled $\lambda = 0$ for the initial reactant and $\lambda = 1$ for the final product. A Rayleigh line is tangent to the product Hugoniot at the CJ point; and the intersection of the Raleigh line and the reactant Hugoniot is marked VN, the von Neumann spike. If the reaction process is very fast, according to classical steady detonation theory, the state would jump from the initial condition O to VN and then move down to CJ along the Rayleigh line. In fact, for an instantaneous reaction as assumed in programmed burn, we do not even concern ourselves with the pathway from VN to CJ, and the product begins to expand at CJ as soon as the detonation wave arrives. That is why we do not see the VN spike when we use programmed burn.

The second part of our discussion involves a partially reacted Hugoniot, labeled $\lambda < 1$. This Hugoniot should be between those of the reactant and the product but much closer to the product one because of the large amount already reacted as seen in the reaction zone, Fig. 4. The intersection of this Hugoniot and the Rayleigh line is labeled "Effective CJ." Because the first phase of the reaction is quite fast with a process time of 5 ns, the pathway coming down from VN follows very

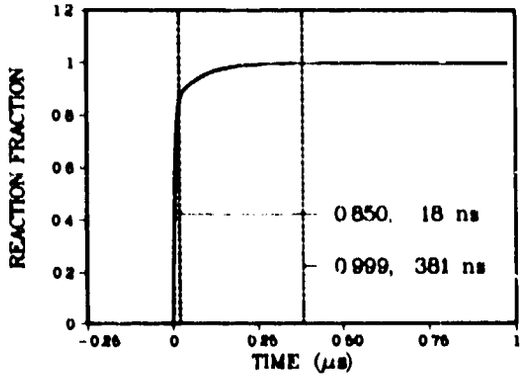

FIGURE 4
PBX 9502 reaction zone showing fast and slow reaction zone thicknesses.

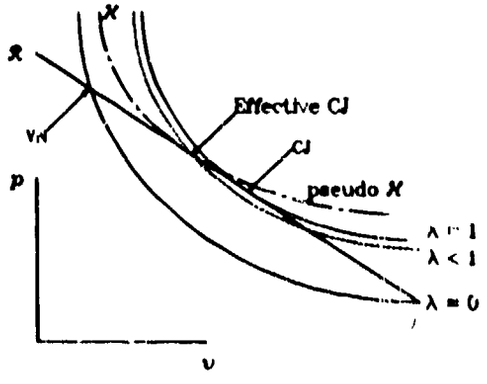

FIGURE 5
Evolution of a pseudo Hugoniot based on an effective CJ state.

closely to the Rayleigh line until it reaches the effective CJ point. The second phase of the reaction is, however, much slower than the first, with a process time of 75 ns. This slow reaction forces the reaction pathway to deviate from the Rayleigh line, starting the expansion from the effective CJ point in a way similar to the classical expansion from the normal CJ state along the principal isentrope. The difference is that the composition of the expanding gas through the effect of λ is still changing, albeit slowly. From the hydrodynamic point of view, the expansion begins at the effective CJ point, not the normal CJ; and, as we can see, the effective CJ pressure is greater than the normal CJ. That is why the inclusion of a slow reaction would result in higher CJ pressure, a condition so vividly illustrated in plate push experiments.[1]

Let us suppose we have no knowledge of the reaction process - in particular, the slow stage - and proceed to construct an EOS based on the measured quantity of "CJ" pressure. The resulting Hugoniot is shown with the label "pseudo". This Hugoniot satisfies the classical detonation, the tangency, requirement. As long as we do not do anything extraordinary and the form of the EOS is well-behaved, the Hugoniot must have the properties we described earlier for a Hugoniot with higher CJ pressure, seen in Fig. 3. It appears more energetic than the real Hugoniot in the low- pressure region as demonstrated in plate push experiments when Al is used, but less so in the high-pressure region. The same conclusion can be reached about the principal isentrope. This pseudo Hugoniot, unfortunately, forms the basis for the construction of EOS in many cases.

So far we only have direct experimental evidence showing the deficiency of the EOS in the low-pressure regime. Still we do not have a concrete result to demonstrate the weakness in the high-pressure domain. However, from the energy consideration involving carbon coagulation, we expect the slow reaction mass fraction should be below 0.1. Instead, a value of 0.15 is used. As Figure 2 shows, the HE experiences a pressure condition over 60 GPa, a consequence of shock reflection from the Ta plate. The magnitude is twice the value of the quoted normal CJ pressure of about 30 GPa. Even reflection from the Al plate can reach a pressure over 40 GPa. The seemingly higher value required for the slow reaction mass fraction is quite likely a manifestation compensating for the effect of less energy of the EOS in the pressure region above CJ. Some overcompensation is seen in a 13-mm PBX 9502 pushing Al plate as a result of a slightly larger slow reaction mass fraction.[1]

The above discussion is based entirely on a quasi-steady concept, but in reality, the pathway is much more complex. For one thing, the Rayleigh line is not fixed; it depends on how the system is driven. The effective CJ, shown in Fig. 5, represents a self-supported asymptotic limit. Therefore, the effective CJ state is not unique with a condition demonstrated clearly in plate push experiments showing the change of the initial velocity jump with respect to varying HE thickness.[1]

4. CONCLUSIONS

For a period of more than 350 nanoseconds after the onset of detonation, the HE is still in a partially reacted state due to the presence of a slow reaction. A construction of the EOS without recognizing the fact automatically assumes it to be the complete product and leads to a pseudo EOS with the deficiencies described earlier. Because of the finite reaction involved for a rather long period, EOS as presented generically in Eq. (4) has a time-dependent character through λ and consequently results in time-dependent, nonsteady detonation, a fact that has been observed for some time. To obtain a true product Hugoniot, the condition must be maintained long enough to reach the quasi-steady state over a wide range of pressure conditions. Construction of EOS based on the false CJ condition alone should be avoided.

ACKNOWLEDGMENTS

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