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Title: A Recipe for implementing the Arrhenius-Shock-Temperature State Sensitive WSD (AWSD) model, with parameters for PBX 9502

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A Recipe for implementing the Arrhenius-Shock-Temperature State Sensitive WSD (AWSD) model, with parameters for PBX 9502

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A reactive flow model for the tri-amino-tri-nitro-benzene (TATB) based plastic bonded explosive PBX 9502 is presented. This newly devised model is based primarily on the shock temperature of the material, along with local pressure, and accurately models a broader range of detonation and initiation scenarios. The equation of state for the reactants and products, as well as the thermodynamic closure of pressure and temperature equilibration are carried over from the Wescott-Stewart-Davis (WSD) model[7,8]. Thus, modifying an existing WSD model in a hydrocode should be rather straightforward.

I. INTRODUCTION AND MOTIVATION

This report presents a modified Wescott-Stewart-Davis model for PBX 9502. Although the original WSD calibration[7] performs well at fitting both shock-to-detonation transition and detonation propagation, there exist several issues that can be improved upon. These are namely:

1. The original WSD model has a sub-micron scale in its first rate term, which necessitates extremely fine grids (≈250 nm) to yield proper asymptotic convergence in numerical solutions. James Quirk, Mark Short[5] and I independently verified this to be the case (likewise Ash Kapila et al[3] had earlier identified this behavior in Ignition and Growth's ZND structure.)

2. Multiple shocks or isentropic compression have a tendency to not be handled well by the original WSD[7] nor the version with desensitization[8]. WSD is generally observed to be too reactive when compared to experiments[1].

3. Changes to pressing density or initial temperature result in changes to sensitivity in the *opposite* direction from experimental observations.

Many of these shortcomings are related to the Ignition and Growth model that WSD utilized. Issue 1 above is related to the "initiation" rate[7], which has a very large rate constant, k_I, of $10^5 \mu s^{-1}$, which subsequently is reduced by a couple orders of magnitude near 3% reaction. The other issues are related to the local pressure sensitivity of the rate law. Essentially, when the pressure becomes large, in either an isentropic or multi-shock scenario, there is nothing limiting the rate. A desensitization model was appended onto WSD in the 2006 Detonation Symposium paper[8], but with few experiments to go by, it wasn't successfully calibrated for many scenarios[1] (but did yield "dead zones" for the examples studied), and would nonetheless still be problematic with respect to Issue 3.

The outline of the remainder of the paper is as follows. Section II details the reactants equation of state (EOS), followed by section III detailing the products EOS. The functional forms describe how the EOSs were actually implemented in Brad Wescott's code, which sometimes differed from the presentation in the original paper[7]. Finally, section IV gives the AWSD rate model and associated constants for PBX 9502, specifically for lots of "recycled" material (lots LANL 79-04 and HOL85F000E-136). Other lots, specifically virgin material, such as HOL80L890-007 and HOL88H891-008 will undoubtedly require adjustment of some rate parameters, as, at least their detonation propagation characteristics are measurably different[9,10].

II. EQUATION OF STATE OF REACTANTS

The WSD model utilizes a "Davis reactants" EOS. This is a thermally complete Mie-Grüneisen equation of state based off a reference isentrope. Following Wescott, et al[7] for the reactants EOS, the relations between specific internal energy, e_r, pressure, p_r, and density, ρ, for the reactants (thus the subscript r) are given by the standard Mie-Grüneisen form:

$$e_r\left(p,\rho\right) = e_r^s(\rho) + \frac{p - p_r^s(\rho)}{\rho\Gamma_r(\rho)} \tag{1}$$

and the inversion of this to obtain

$$p_r\left(e,\rho\right) = p_r^s(\rho) + \rho\Gamma_r(\rho)(e - e_r^s(\rho)). \tag{2}$$

where the reference isentrope, denoted by a superscript s, of the reactants is given by:

$$p_r^s\left(\rho\right) = \begin{cases} \hat{p}\left[\exp(4By) - 1\right], & \rho < \rho_0 \\ \hat{p}\left[\sum_{j=1}^{3} \frac{(4By)^j}{j!} + C\frac{(4By)^4}{4!} + \frac{y^2}{(1-y)^4}\right], & \rho \geq \rho_0 \end{cases} \tag{3}$$

and where $y = 1 - \rho_0/\rho$ and $\hat{p} = \rho_0 A^2/4B$. Here, A is the bulk sound speed of the material, B can be related to the derivative of the bulk modulus at zero pressure,

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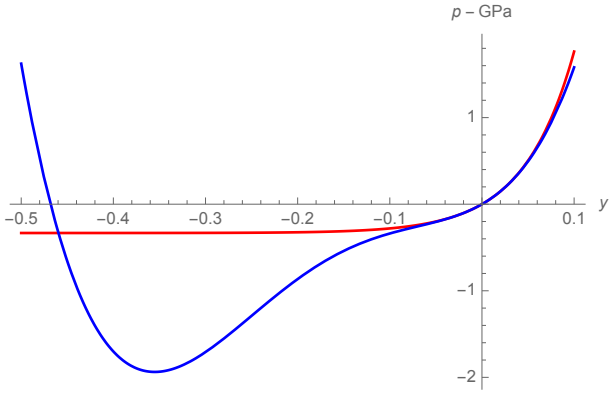

FIG. 1. Isentropes from equation 3 with parameters from Table I. Red curve is for $\rho < \rho_0$, blue curve for $\rho \geq \rho_0$. Note the non-monotonic behavior of the blue curve for $\rho < \rho_0$.

C helps determine the behavior at high shock strengths, and ρ_0 is the reference ambient density. Note that the above two functional forms are continuous up to y^3 at $\rho = \rho_0$. Furthermore, the above $p_r^s(\rho)$ form is monotonically increasing of ρ. *If* no switch is employed (and $C > 0$), and the second term is used for all ρ values, there would always come a point for low enough density where $\frac{dp_r^s}{d\rho} < 0$, implying an imaginary sound speed. Given this fact, the switch in the above equation is required to maintain reasonable mathematical behavior when $\rho < \rho_0$. An example plot of both above functions is shown in figure 1.

From the thermodynamic identity $de = Tds - pdv$, we have for an isentropic process simply $de = -pdv$. Upon substitution of specific volume, v, in favor of density, we obtain $de = \frac{p}{\rho^2} d\rho$. The above reference pressure along the isentrope can then be integrated to obtain the reference energy along the isentrope:

$$e_r^s(\rho) = \int_{\rho_0}^{\rho} \frac{p_r^s}{\bar{\rho}^2} d\bar{\rho} + E_0 \tag{4}$$

where E_0 is integration constant, equivalent to the stored chemical potential energy of the explosive (FLAG likely accounts for this energy offset by other means). The Grüneisen parameter is taken to be:

$$\Gamma_r(\rho) = \begin{cases} \Gamma_r^0, & \rho < \rho_0 \\ \Gamma_r^0 + Zy, & \rho \geq \rho_0 \end{cases} \tag{5}$$

where Z is a constant used to describe the changes to Γ_r with respect to density. Note that the $\rho < \rho_0$ branch is not mentioned in the WSD references, but exists in FLAG. Note also that $y = 0$ at $\rho = \rho_0$, $y \to 1$ as $\rho \to \infty$ and $y \to -\infty$ as $\rho \to 0$.

The reactants temperature, T_r, after a series of manipulations[7], can be obtained as a function of energy and density:

TABLE I. Davis reactants EOS parameters for PBX 9502

Parameter	Value	Unit
A	1.80	mm/μs
B	4.6	
C	0.34	
ρ_0	1.890	g/cm^3
Z	0.0	
Γ_r^0	0.56	
E_0	4.115	kJ/g
C_{vr}^0	0.001074	kJ/g K
α_{ST}	0.4265	
T_0	297	K

$$T_r(e,\rho) = T_r^s(\rho)\left\{\frac{1+\alpha_{st}}{C_{vr}^0 T_r^s(\rho)}\left[e - e_r^s(\rho)\right] + 1\right\}^{\frac{1}{1+\alpha_{st}}} \tag{6}$$

where $T_r^s(\rho)$ is the temperature along the reference isentrope:

$$T_r^s(\rho) = \begin{cases} T_0\left(\frac{\rho}{\rho_0}\right)^{\Gamma_r^0}, & \rho < \rho_0 \\ T_0 \exp(-Zy)\left(\frac{\rho}{\rho_0}\right)^{(\Gamma_r^0 + Z)}, & \rho \geq \rho_0, \end{cases} \tag{7}$$

Here, C_{vr}^0 is the reactants specific heat at constant volume at the reference temperature; α_{st} determines how the specific heat changes with respect to temperature and T_0 is the reference temperature.

Other thermodynamic manipulations can be performed to explicitly determine entropy. But since entropy isn't used in either the WSD or AWSD rate model, it is not discussed here. Again, further details can be found in the references.

For the reactants EOS, the following parameters are required to define the model: A, B, C, ρ_0, Z, Γ_r^0, E_0, C_{vr}^0, α_{st} and T_0. A representative set of parameters, recently calibrated[11], for PBX 9502 are given in Table I.

III. EQUATION OF STATE OF PRODUCTS

The Davis functional form for detonation products is also given by a standard Mie-Grüneisen form:

$$e_p(p,\rho) = e_p^s(\rho) + \frac{p - p_p^s(\rho)}{\rho \Gamma_p(\rho)} \tag{8}$$

and

$$p_p(e,\rho) = p_p^s(\rho) + \rho \Gamma_p(\rho)(e - e_p^s(\rho)), \tag{9}$$

where the subscript p denotes the products EOS. Following[7] and references therein, the pressure on the

principal Chapman-Jouguet (CJ) isentrope is given by:

$$p_p^s(\rho) = p_c \frac{\left[\frac{(\rho v_c)^{-n}}{2} + \frac{(\rho v_c)^n}{2}\right]^{a/n}}{(\rho v_c)^{-(k+a)}} \frac{k-1+F(\rho)}{k-1+a} \tag{10}$$

where

$$F(\rho) = \frac{2a(\rho v_c)^n}{(\rho v_c)^{-n} + (\rho v_c)^n}. \tag{11}$$

Given $p_p^s(\rho)$, one can integrate $de = \frac{p}{\rho^2} d\rho$ to obtain the energy along the isentrope:

$$e_p^s(\rho) = e_c \frac{\left[\frac{(\rho v_c)^{-n}}{2} + \frac{(\rho v_c)^n}{2}\right]^{a/n}}{(\rho v_c)^{-(k-1+a)}}, \tag{12}$$

where

$$e_c = \frac{p_c v_c}{k-1+a}. \tag{13}$$

The Grüneisen parameter is given by:

$$\Gamma_p(\rho) = k - 1 + (1-b)F(\rho). \tag{14}$$

The parameters p_c, v_c, a, k and n define the principal CJ isentrope. Once these five parameters are determined, the parameter b sets the Γ_p and thus the off-isentrope behavior. Overdriven Hugoniot data is often employed to obtain off-isentrope behavior.

The temperature of the products is given by:

$$T_p(e,\rho) = T_p^s(\rho) + \frac{e - e_p^s(\rho)}{C_{vp}}, \tag{15}$$

where the temperature on the principal isentrope is give by:

$$T_p^s(\rho) = T_c \frac{\left[\frac{(\rho v_c)^{-n}}{2} + \frac{(\rho v_c)^n}{2}\right]^{(a/n)(1-b)}}{(\rho v_c)^{-(k-1+a(1-b))}}, \tag{16}$$

where

$$T_c = \frac{2^{-ab/n}}{k-1+a} \frac{p_c v_c}{C_{vp}}. \tag{17}$$

A set of PBX 9502 products parameters are given in table II. In addition to the reactants and product EOSs, a closure rule is needed when the material is partially reacted. A common closure rule is to assume that the pressure and temperature are equilibrated between reactants and products. This is assumed in WSD and AWSD. It is likely not the most realistic closure model, but is a thermodynamically viable pathway. Further discussions of potential closure rules are discussed in Matignon, et al[4].

TABLE II. Davis products EOS parameters for PBX 9502

Parameter	Value	Unit
a	0.798311	
k	1.35	
v_c	0.75419	cm^3/g
p_c	3.2	GPa
n	2.66182	
b	0.58	
C_{vp}	0.001072	kJ/g K

IV. THE AWSD RATE LAW

After much examination of experiments under a wide range of initial temperatures, as well as complex loading scenarios, it was found that a shock-like temperature in conjunction with local pressure matches the available data with reasonable fidelity. It is undoubtedly not the only means to fit the given data, but appears to work the vast majority of the time. Local pressure is a common state variable used in Ignition and Growth and the original WSD formulations, and is readily available since it is used in hydrodynamic computations. On the other hand, the shock temperature is not often tracked in hydrodynamic simulations. For the WSD model, the local temperature is utilized in the thermodynamic closure of pressure and temperature equilibration between reactants and products in the reaction zone. So, given the local temperature and other thermodynamic state variables, one needs to construct an approximation to the shock temperature in some reasonable fashion. Prior to discussion of how to compute the shock temperature for AWSD, the next two subsections review what is currently performed by WSD.

A. How the shock state is currently determined in WSD

It should be noted that the current WSD model does track the shock state, specifically the shock density, which in turn is used in determining whether the HE is in the "shock initiation" or "detonation propagation" region. The bulk of the WSD rate sensitivity, though, comes from examining the local pressure. The computational methodology for determining the shock density, ρ_{SH}, is determined by examining a combination of the local density, ρ, and the local reaction progress variable, λ. Specifically, ρ_{SH} is set by:

$$\rho_{SH} = \begin{cases} \max\left(\text{current } \rho_{SH}, \rho\right), & \text{if } \lambda < \lambda_c \\ \text{current } \rho_{SH}, & \text{if } \lambda \geq \lambda_c. \end{cases} \tag{18}$$

For Lagrangian algorithms, the above is simply performed in every cell during every timestep. For an Eulerian scheme, in addition to this, one needs to advect this quantity with the local flow speed, as one would do

for other passively advected quantities. In current implementations, $\lambda_c = 0.05$. There is some insensitivity to the choice of λ_c, since under detonation conditions the density has a relative maximum at the shock point. For other scenarios, such as isentropic compression, or shock/ramp loading, it becomes a bit nebulous as to how to choose the proper shock state. It should also be pointed out that the length between the shock and $\lambda = 0.05$ for a ZND CJ detonation in PBX 9502 is approximately 3 μm, which is typically not resolved in hydrodynamic simulations. Nonetheless, this is the current recipe for determining the shock state in WSD.

B. WSD desensitization rate

Furthermore, the WSD implementation in flag also has a desensitization "rate"[8]. This rate is associated with the "rate of removal of potential hot spot sites" and is a function of pressure:

$$\dot{\psi} = kpH(p - p_{HEL})[1 - H(p - p_{max})] \qquad (19)$$

This desensitization rate turns on once the pressure reaches the Hugoniot Elastic Limit, p_{HEL}, (i.e. roughly where plastic deformation starts to remove void space), and ends at the point where p reaches p_{max}. Here, $H(x)$ is the Heaviside function, $H(x < 0) = 0$ and $H(x > 0) = 1$. The rate is also proportional to the local pressure, so that higher pressures lead to faster removal of hotspots. For PBX 9502, the WSD model with desensitization[8] used $p_{HEL} = 0.07$ GPa, which is consistent with Dick et al[2] reported value of 0.073 GPa and $p_{max} = 6$ GPa, which is slightly below the lowest shock to detonation pressure observed in PBX 9502. Here, ψ starts out at zero, and when $\psi = 1$, the hotspots are turned off. Turning off the main WSD rate is accomplished by premultiplying the WSD rate law by $H(1-\psi)$. So, when $\psi > 1$, the WSD rate is stopped.

C. Approximating the shock temperature for AWSD

As mentioned previously, the AWSD state sensitivity involves knowledge of the temperature of the material as it passes through a shock. Unlike the density field, which has a relative maximum at the shock during detonation, the temperature generally continues to increase as it approaches the fully burned state. For the EOS parameters presented here, see figure 2 for the post shock temperature as a function of reaction progress for several initial temperatures. Thus, a different algorithm must be utilized to approximate the shock temperature.

A function which has a relative maximum at the shock, and equal to the shock temperature at $\lambda = 0$, is:

$$T^*(T, \lambda) = T\left(1 - a_T \exp\left(T_c/T\right)\lambda\right) \qquad (20)$$

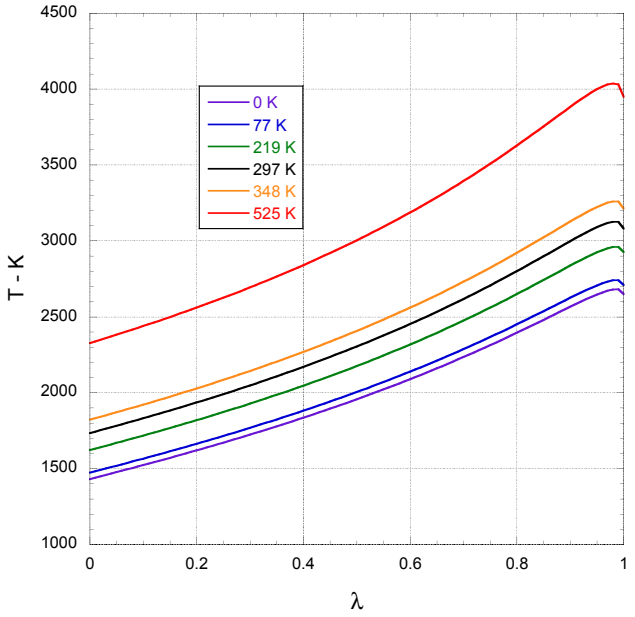

FIG. 2. Temperature through the reaction zone for a steady ZND wave at the CJ velocity for material at the initial temperatures given in the key.

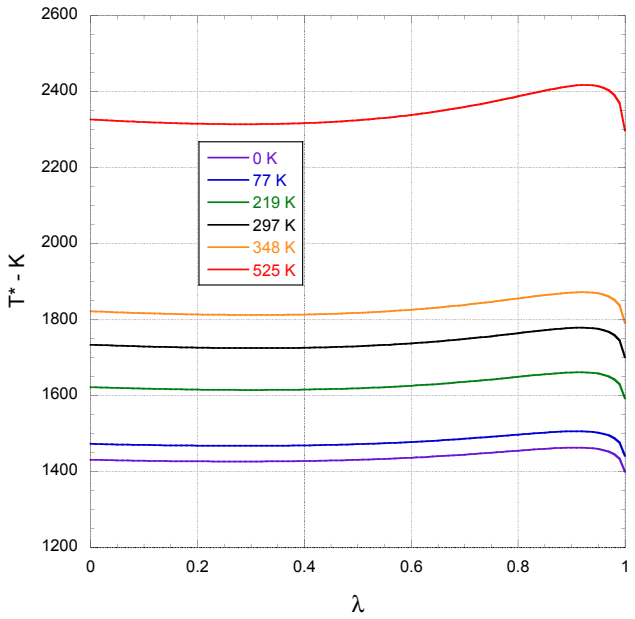

FIG. 3. T^* through the reaction zone for a steady ZND wave at the CJ velocity for material at the initial temperatures given in the key.

where a_T and T_c are constants. These constants are determined by examining the temperature through the reaction zone, i.e. $T(\lambda)$, for a variety of initial material temperatures. For $a_T = 0.327$ and $T_c = 971$ K, figure 3 shows the function T^* through the ZND CJ reaction zone. Note that the function has a relative maximum at the shock state and is very flat in the range $0 < \lambda < 1/2$.

To compute the shock temperature over a wider range of initial conditions (beyond simply examining ZND CJ detonation waves), it is necessary to limit the query of T^* to the vicinity near the shock. This is accomplished through the use of a simple "timer" rate. Specifically, we integrate

$$\dot{\zeta} = k_\zeta H(p - p_\zeta) \tag{21}$$

Here, once p reaches p_ζ, the rate proceeds at $\dot{\zeta} = k_\zeta$. From experiments[6] on LX-17, an explosive similar to PBX 9502, it was determined that shock pressures between 0.52 and 0.68 GPa were sufficient to compress out voids in the material, so the value $p_\zeta = 0.6$ GPa is chosen. We don't have data on what timeframe it takes to compress the voids out of the porous explosive, which would ideally be needed to determine k_ζ. A reasonable choice of $k_\zeta = 20\ \mu\text{s}^{-1}$ is made, thus material that has been compressed above 0.6 GPa will reach $\zeta = 1$ after 50 ns. If further data becomes available, this rate can be revisited.

The final computed approximate shock temperature is then determined by

$$T_{SH} = \begin{cases} \max\left(\text{current } T_{SH}, T^*\right), & \text{if } \lambda < \frac{1}{2} \text{ and } \zeta < 1 \\ \text{current } T_{SH}, & \text{otherwise.} \end{cases} \tag{22}$$

In general, for quiescent material, T_{SH} should be set to the ambient material temperature. Likewise, for quiescent material, ζ should be set to zero.

D. The AWSD rate law

The current AWSD rate law, as mentioned previously, relies on an approximation to the shock temperature. Additionally, the local pressure, p, plays a role as well as the reaction progress, λ, as:

$$\frac{D\lambda}{Dt} = R(T_{SH}, p, \lambda), \tag{23}$$

where $\frac{D}{Dt}$ is the total or material derivative. Specifically, the rate is:

$$R = F_p \left(F_1 + F_2\right) F_\lambda \tag{24}$$

where:

$$F_p = \begin{cases} \exp\left(-\left(p_s/p\right)^{n_p}\right), & \text{if } p > p_\zeta \\ 0, & \text{otherwise.} \end{cases} \tag{25}$$

$$F_1 = k_1 \exp\left(-T_1/T_{SH}\right)\left(\lambda + a_1 F_p\right)\left(1-\lambda\right)^{b_1}, \tag{26}$$

$$F_2 = k_2 \exp\left(-T_2/T_{SH}\right)\left(1-\lambda\right)^{b_2} \tag{27}$$

and

$$F_\lambda = f_s + \frac{1}{2}\left(1 - f_s\right)\left(1 - \tanh\left(\frac{\lambda - \lambda_c}{\delta_\lambda}\right)\right). \tag{28}$$

TABLE III. AWSD rate parameters

Parameter	Value	Unit
n_p	0.6472	
p_s	17.70	GPa
k_1	1019	μs^{-1}
T_1	2678	K
a_1	0.1648	
k_2	23330	μs^{-1}
T_2	8091	K
f_s	0.05002	
λ_c	0.8783	
δ_λ	0.01445	

There are several qualities to point out about the above rate. Firstly, the pressure and shock temperature sensitivities are multiplicative, so if either are particularly low, the resulting rate is also low. The F_p rate has a functional form that is known as a stretched exponential (assuming $n_p < 1$). In F_1, the temperature portion is Arrhenius in the shock temperature, with an "activation temperature" of T_1. The rather unconventional functional form for the λ dependence stems from the ignition and growth-like concept, without having to deal with some of the singular behavior of the original forms; $b_1 = 2$ was fixed to yield a shock-to-detonation buildup curve similar to what is observed experimentally. In F_2, there is a more traditional depletion form (here the chosen depletion factor of $b_2 = 0.9$ was simply a choice to force the reaction zone to not be infinitely long.) In F_2, there is a separate "activation temperature", T_2. F_λ is a very simple way to lower the rate from its nominal value by a factor of f_s, where F_λ transitions from near unity to f_s, centered at $\lambda = \lambda_c$, with a characteristic scale of δ_λ.

Here, there are a total of 10 free rate parameters. For recycled lots of PBX 9502, see table III for the calibrated rate parameters. Discussion of the calibration procedure is discussed in a separate manuscript[12].

V. ACKNOWLEDGEMETS

I would like to thank Matt Price and James Quirk for carefully reviewing this manuscript.

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