Temperature-based model for condensed-phase explosive detonation

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Abstract. Simple reactive flow models for condensed explosives have four requirements: two equations of state (EOS), one for the unreacted condensed-phase explosive and one for its detonation products, a reaction rate law that converts the explosive in products and a mixture rule to compute the biphasic partially reacted states (with both unreacted explosive and products). Generally, the chemical reaction rates are governed by local temperature. Nonetheless, temperature fields are scarcely known, especially in detonating explosives. Hence this quantity is not provided by the usual unreacted explosive EOS with the required accuracy. As a consequence, for shock initiation and detonation phenomena, rate laws are based on easily measurable properties such as pressure, density, compression or particle velocity. In this work, we try to build a temperature-based reaction rate law. This model is expected to give interesting results as regards shock initiation and desensitization while remaining accurate for detonation propagation.

1. Introduction
Precise modelling of detonation is of great importance to predict phenomena such as initiation, shock desensitization or dead zone in corner turning experiments. Most of the reactive flow models dedicated to condensed-phase explosives are made of five items. 1) An EOS of the unreacted explosive which is function of either energy and volume \( p(e, v) \) or pressure and volume \( e(p, v) \). Temperature, when given, is generally computed from constant or linearly dependant \( C_v \) models. 2) An EOS of the burnt phase. The same conclusions as regards temperature as above can be drawn for simple EOS as JWL or Davis’ EOS [1, 2]. For burnt phase EOS computed by means of thermochemical codes [5, 6], thermodynamical quantities are accurately known provided an appropriate calibration on relevant data is performed. 3) A closure law of the unreacted/reacted system. This law can be an equilibrium condition on pressures and volumes of the two phases or on pressures and temperatures. In the CREST reactive burn model, the closure law is made of a pressure equilibrium and an isentropic evolution of the unburnt phase [3, 4]. 4) A mixture rule to compute properties of partially reacted states. An ideal mixture rule is almost always used: \( e = \lambda e_p(p_p, v_p) + (1 - \lambda)e_s(p_s, v_s) \) (where \( \lambda \) stands for the burnt fraction, quantities subscripted \( s \) stand for unreacted solid phase and quantities subscripted \( p \) stand for detonation products). 5) A reaction rate law which is usually function of pressure and density: Forest-Fire [7], Johnson-Tang-Forest [8, 9], Lee-Tarver [10], Wescott-Stewart-Davis [11] among others. These reaction rate laws try to reproduce the chemical reactivity occurring in the reaction zone. But pressure and density are not the relevant quantities to describe chemical...
phenomena such as decomposition reactions of explosives. On the contrary, relevant quantities could be the entropy deposited by the shock or a local temperature.

Lambourn and James [12] have analyzed a large amount of recent in-material gauge results from PBX 9502. Significant correlations have been highlighted: a monotonic relationship between the local shock strength and the time-to-peak particle velocity along each particle path and the simple scaling of velocity histories along the particle path that exists at a common local shock strength from shots with different initial conditions. These correlations are also valid for double shocks or thin pulses. The authors conclude that a strong relationship occurs between the reaction rate and the local shock strength, and that shock entropy is the most relevant measure of that shock strength.

The recent CREST reactive burn model [3, 13] is based on the analysis of Lambourn and James: the reaction rate is a function of the entropy of the unreacted explosive. This model is shown to be very accurate on single-shock, double-shock and thin-pulse shots on the HMX-based explosive EDC37 [3], without any bolt-on desensitization model. That natural desensitization capability ensues from the quasi isentropic evolution of shock entropy (and shock temperature) after a second shock. Accuracy on detonation failure diameter has also been proven by Whitworth [14]. Moreover Lambourn and James [15] suggest that a second-order evolution term based on temperature is likely to be used.

2. WSD(T) temperature-based model
The well-known WSD reactive flow model [11] is largely used particularly because the reaction rate is very easy to handle. The rate is made of four regimes covering ignition (called I in [11]), growing of the ignition (IG) and established detonation (DG and B). The transition between the different regimes is performed by switching functions (depending on burnt fraction and shock density). The ignition regime (I) is function of the density and the burnt fraction. The IG, DG and B regimes, functions of pressure, are written:

\[ r_X = k_X \left( \frac{P}{P^*} \right)^{n_X} f_X(\lambda) \]

where \( X \) stands for IG, DG or B, and \( f_X(\lambda) \) are functions of the burnt fraction. These regimes can be calibrated separately on specific data such as experimental run-to-detonation distance, celerity-curvature curve or the critical failure diameter. As a consequence, the reaction rate is a function of pressure, density and shock density. We propose in this work to build a reaction rate, called WSD(T) hereafter, by replacing the pressure terms by similar temperature terms:

\[ r_X = k_X \left( \frac{T}{T^*} \right)^{n_X} f_X(\lambda) \]

All other terms are kept identical, that is the ignition rate \( r_I \) is still function of density and the switching functions are not modified. Further modifications of theses terms will be the subject of future work.

3. Calibration of the WSD(T) model
Calibration of the WSD(T) model is performed for PBX 9502. EOS and parameters of unreacted explosive and detonation products are taken from [11]. In our case, the closure law is a pressure and volume equilibrium [16].

As described in [11], calibration of ignition terms is performed by means of time-dependent, one dimensional numerical simulations of the shock initiation experiment. The parameters \( T^*, k_I, n_I, k_{IG}, n_{IG} \) and \( a \) are adjusted to match the experimental run-to-detonation distance
Table 1. Parameters of WSD(T) model for PBX 9502.

| Parameter | Value       |
|-----------|-------------|
| $a$       | 0.21935     |
| $T^*$     | 1350 K      |
| $k_I$     | $1.025 \times 10^5$ $\mu s^{-1}$ |
| $n_I$     | 7.175       |
| $k_{IG}$  | 30.75 $\mu s^{-1}$ |
| $n_{IG}$  | 7.175       |
| $\rho_C$  | 2710 kg.m$^{-3}$ |
| $k_{DG}$  | 68.7 $\mu s^{-1}$ |
| $n_{DG}$  | 0.74        |
| $k_B$     | 0.50 $\mu s^{-1}$ |
| $n_B$     | 1.5         |

versus shock pressure data. Results are summarized in table 1 (upper part). Simulated run-to-detonation distances with WSD(T) are in very good agreement with both WSD and experimental results (figure 1). Parameters included in the $r_I$ ignition term (not modified) are very close to the values of [11]. Nonetheless this regime has very little influence since it vanishes for burnt fractions above 0.025. This regime only serves to initiate decomposition while the main regime involved in the shock initiation experiment is the IG regime.

The two remaining terms ($r_{DG}$ and $r_B$) are calibrated against experimental detonation shock speed ($D_n$) - shock curvature ($\kappa$) relationship. The $D_n(\kappa)$ relation is computed by using a shooting method to integrate the reaction zone from the shock front to the sonic surface (where both sonicity and thermicity vanish) [17]. The $\rho_C$ parameter is set to ensure the experimental failure diameter and the turning point at large curvatures in the $D_n(\kappa)$ plot. The results obtained are reported in table 1 and on figure 2. WSD and WSD(T) are in good agreement.

4. Shock initiation
For porous explosives, like PBX 9502, the density of the initial state is of great importance. Indeed, without any consideration of reactivity, if the density of the explosive is lowered (porosity increased), the shock pressure reached for the same material speed will be lower since energy is dissipated to close the porosity. It is also obvious that temperature of the explosive will be higher. Such a behavior is easily obtained by changing only the initial density of the EOS of the unreacted explosive. But as a consequence of the evolution of pressure, a pressure-dependent reaction rate is not able to reproduce the increase of sensitivity with increase of porosity, as can be seen on figure 3. WSD model predicts the contrary: a decrease of sensitivity with increasing porosity. The proposed WSD(T) model, function of temperature, naturally reproduces the increase of sensitivity with porosity (figure 3).

5. Desensitization
Recently, Aslam et al. [18] proposed an experimental approach that can be used to gain information on the unreacted Hugoniot by multi-shock compression. An impactor made of Tantalum (2.5 mm thick) and TPX (3 mm thick) inserted in a sabot in Lexan impacts a target made of PBX 9502 (6 mm thick), followed by a LiF window. With such a device, a first weak shock propagates in the target followed by a second strong shock. Due to desensitization by the first weak shock, PBX 9502 is not supposed to react when subject to the second strong shock. If we suppose no reactivity of PBX 9502, the shocked state of the explosive at the PBX 9502/LiF interface is as plotted in figure 4 (lines). Up to four successive shocks can be discretized (especially the first weak shock and the second strong one).
If reactivity is assumed as in WSD model, desensitization does not occur. As can be seen in figure 4 (dashed lines), the first shock is not strong enough for PBX 9502 to react completely and immediately (dashed blue line), but the explosive does decompose and a complete reaction happens just before the arrival of the second shock at 1.7 \( \mu s \). With the WSD(T) model (dot-dashed lines), the first shock is non-reactive. Decomposition starts after the second shock (dot-dashed blue line) but with a very small slope (\(~0.1 \mu s^{-1}\)). Fast and complete decomposition occurs around 0.7 \( \mu s \) after the second shock. Although WSD(T) does not predict global desensitization of the explosive, it does predict a drastic reduction of the reactivity with multi-shock compression. To our opinion, this is a great improvement over WSD. Additional modifications of the remaining density-dependent term of the reaction rate are expected to improve predictions of desensitization.

6. Conclusions
It has been shown that temperature of an unburnt shocked explosive is a good candidate to drive rate laws of decomposition of explosives. Starting from the well known WSD model, a new reaction rate is proposed. This new rate is made of four regimes covering ignition, growing of the ignition and established detonation. Transition between the different regimes is performed by switching functions. The ignition regime is function of density and burnt fraction. The three other regimes are functions of shocked temperature and burnt fraction. This new reaction rate has been calibrated for PBX 9502 against experimental data of the literature. Instead of WSD model, this new WSD(T) model reproduces the evolution of sensitivity of explosives with porosity. WSD(T) also predicts a drastic reduction of the reactivity in the case of multi-shock compression without any bolt-on desensitization model. Modification of the density-dependent terms in the WSD(T) model and rate laws based hot spot models [19, 20] are subject to future work.
Figure 3. Run-to-detonation distance vs input shock pressure. Black line is a fit of experimental data provided in [11], blue curves stand for original WSD model, red curves stand for the WSD(T) model. Lines correspond to the nominal 1895 kg.m$^{-3}$ density while dashed lines correspond to an initial density of 1800 kg.m$^{-3}$. Arrows indicate the increase of porosity.

Figure 4. Shocked state of PBX 9502 at the PBX 9502/LiF interface for a flyer velocity of 2139 m.s$^{-1}$. Pressure (P) is in black, temperature (T) in red and burnt fraction ($\lambda$) in blue. Lines correspond to non-reactive explosive, dashes stand for WSD and dot-dashes stand for WSD(T).

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