Multiple shock compressions in tri-amino-tri-nitro-benzene based energetic materials

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Abstract. Various hazard and vulnerability scenarios for energetic materials involve multiple shock compression. Triaminotrinitrobenzene (TATB) based explosives are comparatively insensitive to shocks of low impacts and therefore considered suitable materials for the devices vulnerable to low shock impacts. Multiple shock compression may further desensitize these materials. Although the desensitization criterion for HMX based plastic bonded explosive PBX9404 has long been determined, no such criterion for TATB based explosives has yet been determined. In the present paper the desensitization criterion for TATB based explosive is proposed, based on recently performed experiments. By using a model which employs the proposed criterion to account for the desensitization effects in TATB based explosives, numerical simulations of the multiple shock experiments have been performed. The calculated results agree closely with the experimental results.

1. Introduction
TATB based explosives belong to the class of insensitive explosives. These explosives have been of great interest because of their remarkable insensitivity and relative invulnerability to accidental shocks. The explosives LX-17 (92.5% TATB/7.5% kel-F 800) and PBX9502 (95% TATB/5% kel-F 800) have been extensively studied and show non ideal behaviour. Their failure diameters are comparatively large [1-2] and detonation waves are more severely affected by abrupt geometry changes than other explosives [3]. Their detonation and shock initiation properties have stronger dependence on the initial temperature and particle size distribution [2, 4]. Unlike other explosives, the detonation waves in unconfined explosives fail even at 97% of their Chapman-Jouguet (CJ) velocities [1]. The shock initiation threshold criterion $P^2\tau$ has been observed to hold for many explosives [5]. However, Honodel et al. [6] noticed the departure of TATB explosives from $P^2\tau = constant$ criterion at low pressures. His analysis suggested that at low shock pressures TATB based explosives may behave like homogeneous explosives. Souers [7] discussed the initiation thresholds for TATB based explosives. Haskins et al. [8] proposed a modified criterion for the prediction of shock initiation threshold for superfine TATB. It is now an established fact that passage of a shock which is weak enough to detonate the explosive, but strong enough to compress the explosive above it Hugoniot elastic limit (HEL), desensitizes the explosive for the succeeding shock which could otherwise detonate the explosive if the explosive were not preshocked. Desensitization criteria for PBX-9404 has been experimentally determined by Campbell [9] by subjecting PBX-9404 to pre-shocks in the...
pressure range 1.0 to 2.4 GPa. Using the experimental data, a relation was proposed between the preshock pressure \( P \) and the time required \( \tau \) for desensitization of PBX-9404 as \( P^{2.2} \tau = 1150 \) (kbar\(^2\) μs). Unfortunately no such experiments have been performed for LX-17 explosive and consequently no such relation for the explosive is available to date, although the need has been felt by many authors [10, 11]. Recently Vandersall et al. [12] performed shock desensitization and detonation quenching experiments on LX-17 using a Teflon flyer fired from a gas gun to impact at one end of a cylindrical sample of LX-17 and generate a single low amplitude shock wave to pre-shock the explosive sample just prior to detonating it from the other end. However some more experiments are needed to exactly determine the limits of the pressure range and the time window for desensitization. We have proposed a desensitization model for PBX-9404 and applied it for simulation of various preshock desensitization experiments for PBX-9404 in a recent study. In the present study we tried to adapt the desensitization model for LX-17 using the experimental results of Vandersall [12], and simulated the desensitization experiments on LX-17 reported in literature [10, 12, 13]. A brief overview of the modelling of desensitization effects is given in the next section. The available preshock desensitization experiments are simulated in section 3 for validation of the model.

2. Modelling multiple shocking effects

Most of the phenomenological reactive flow models in use today do not have capability to account for multiple shocking effects such as desensitization induced by preshocking. Some special treatments is required in these models to deal with such problems. For example, JTF model [14] accounts for preshock desensitization effects by restricting creation of hot spots to the first shock only, while the second shock is allowed to provide the adiabatic heating only. Wescott-Stewart-Davis (WSD) model [15] has later been improved to a temperature dependent form [16] and is made capable to predict the reduction of the reactivity with multi-shock loading. Although the reactive burn model CREST [17] that utilizes entropy-dependent reaction rates, has been successful updated [18] for predicting the persistent dead zones in corner turning experiments [13] on PBX 9502 explosive at different initial temperatures without any shock desensitization treatment, however, the detail of the improved CREST model for PBX9502 has not been published yet.

We have tried to annex the desensitization model for LX-17 in Lee Tarver ignition and growth model, as it is one of the popular reactive burn models in use today and has been embedded in various commercial hydrocodes. It has been parameterized for a number of explosives and has been used to solve a variety of problems [4, 19-28]. Lee-Tarver model [24, 28] employs separate Johns-Wilkins-Lee (JWL) equations of state (EOS) for both the solid unreacted explosive and the reaction products, and a reaction rate equation. The three term reaction rate equation for the standard model is of the form

\[
d\lambda = R_I + R_{G1} + R_{G2},
\]

where

\[
R_I = t(1 - \lambda)^b(\eta - 1 - C_{crit})^c, \quad R_{G1} = G_1(1 - \lambda)^e p^\gamma \quad \text{and} \quad R_{G2} = G_2(1 - \lambda)^e p^\gamma,
\]

while \( \lambda \) is the fraction reacted, \( t \) denotes the time, \( \rho_0 \) is the initial density, \( \rho \) is the current density, \( C_{crit} \) is the critical compression for ignition and \( P \) is pressure. \( I, b, x, G_1, G_2, c, d, e \) and \( g \) are constants. \( R_I \) is the ignition term which represents the creation of hot spots; while \( R_{G1} \) and \( R_{G2} \) are the growth and completion terms, respectively. The limit for \( R_I \) is from 0 < \( \lambda \) < \( \lambda_{igmax} \), for the \( R_{G1} \) is from \( \lambda_{G1min} < \lambda < \lambda_{G1max} \) while \( R_{G2} \) is calculated from \( \lambda_{G2min} < \lambda < 1 \).

The standard equation (1) has no mechanism to deal with desensitization and thus results rapid reaction by second shock. Several attempts have been made to modify the reaction rate equation (1) to simulate the desensitization experiments. For example, for account for desensitization of LX-17 by reflected shocks, Tarver [29] added a condition in the reaction rate equation that if a critical range of pressures was applied to the explosive, the reaction rate was forced to zero. Whitworth [30] modified
the reaction rate equation by making it dependent on the preshock pressure \( P_{shk} \), in such a way that the pressure \( P = \min (P, P_{shk}) \) was used to find the reaction rate, where \( P \) is local pressure. A qualitative agreement was reported for the calculations of Mulford’s precursor shock experiment [31]. However, contrary to the experimental results for detonation quenching experiments [9], the model gives increased rate of quenching with decreasing precursor shock pressure. The modification proposed by Oliveira [25] introduces the desensitization rate depending upon the degree of desensitization \( \phi \), which varies from 0 to 1. The critical compression \( \alpha \) in the ignition term \( R_I \) is thus modified accordingly. The switching limit of the growth term from 0 to \( \lambda G_{1\text{max}}(\phi) \) was also changed, defined by \( \lambda G_{1\text{max}}(\phi) = \lambda_c \phi \), where \( \lambda_c > 0 \). This modification has some drawbacks mainly due to empirical nature of the parameters and the form of the shape function.

We have proposed a desensitization model which could successfully reproduce the preshock desensitization experiments for an HMX based explosive EDC-37 [32, 33], and could qualitatively reproduce the detonation quenching and reflected shock experiments for LX-17 explosive. This model has been used to simulate the desensitization effects during shaped charge jet penetration in covered explosives [34]. Here we briefly describe the model, the details can be found elsewhere. Our model makes use of the assumption [35] that the preshock shock desensitization takes place for the preshock pressure \( P \) in certain range \( (P_1 < P < P_2) \) specific to the explosive. Based on the experiments, it is also believed that the shock desensitization is a time dependent process. Our model utilizes \( P^n \tau \) desensitization criterion suggested by Campbell [9] to measure the degree of desensitization, where \( n=2.2 \) for PBX 9404. Thus the degree of desensitization \( \phi \) is defined as

\[
\phi = \frac{1}{P^n \tau} \int_{t_1}^{t_2} P^n (t) \, dt
\]  

(2)

where the time \( t_1 \) corresponds to the pressure \( P_1 \) below which it is assumed no desensitization occurring to the explosive and after which the desensitization begins and the time \( t_2 \) corresponds to the pressure \( P_2 \) where the desensitization is maximum, i.e. at \( P=P_1, \phi = 0 \) and at \( P=P_2, \phi=1 \). The critical compression \( C_{crit} \) has to be increased to account for the loss of the potential hot spot generation sites in the desensitized explosive, and is defined as

\[
C_{crit}(\phi) = C_{crit} + \phi C_I,
\]  

(4)

The growth term coefficients are adjusted as follows

\[
G_1(\phi) = (1-\phi)G_1, \text{ and } G_2(\phi) = (1-\phi)G_2
\]

(5)

where \( C_I \) is an empirical constant adjusted for an explosive by comparing the calculation results with the experimental results, to inhibit the ignition term in reaction rate equation (hence inhibiting the detonation in the preshocked explosive).

2. Desensitization criteria for LX-17 and simulation of desensitization experiments

As discussed earlier, Campbell [9] fitted a curve for the pressures and time durations to completely desensitize the PBX9404 explosive samples and found that the points lying on \( P^n \tau \) curve correspond to complete desensitization of the sample. It can be noted that \( n=2.2 \) for desensitization while \( n=2 \) for shock initiation criterion. The TATB based explosive have been observed to significantly deviate [6] from the \( P^n \tau \) shock initiation criteria which many other high explosives follow [5]. Therefore it is quite possible that the desensitization criterion would be different for TATB based explosives. A series of experiments such as those of Campbell [9] is indeed required to find \( n \) in \( P^n \tau \) desensitization criterion. Recently, Vandersall et al. [12] performed a first set of experiments using a gas gun and manganin pressure gauges to study the desensitization effects and calibrated the empirical constants in the Oliveira’s desensitization model. In order to quantitatively measure the interaction of weak shock with the LX-17 detonation waves, Vandersall et al. [12] used a 101 mm gas gun to launch Teflon flyers at low (277 to 491 m/s) velocities to impact LX-17 targets containing embedded pressure gauges to record the pressure histories. The target explosive LX-17 was in the form of thin disks with pressure gauge packages inserted in between, with the total explosive thickness approximately 28 mm. The
target explosive was detonated at the opposite ends at correct times using the detonators and LX-10 boosters. The LX-10 detonation waves promptly initiated LX-17 detonation waves that then travelled through the charges and interacted with the weak nonreactive shock waves caused by flyer impact on the opposite end. Both of the waves interact within the LX-17 targets. The simulation setup for the experiment is shown in Figure 1. The pressure history at different gauge points shows that the impact of Teflon flyer with velocity of 277m/s (shot # 4829) could not desensitize the target, while the impact of flyer with 357m/s (shot # 4817) was able to desensitize the target. These flyer velocities correspond to impact pressures of 0.52 and 0.68 GPa for the time durations of 3.0 and 3.3 µs respectively. There was a third shot also, with flyer velocity of 491 m/s impacting on one side of the target 10 µs earlier than the detonators were fired on the opposite end, however, the gauge record is not reliable probably due to stretching of pressure gauges by the preshock pressure.

By using the user equation of state option [36], we implemented the modified the Lee-Tarver ignition and growth model in LS-DYNA. If the input pressure ($P$) lies within the specific range for desensitization ($P_1 < P < P_2$), the degree of desensitization calculated and stored as a history variable for every computational element and is updated after every computational time step. Owing to the cylindrical symmetry of the problem, two-dimensional axi-symmetric Lagrange grids having 10 elements per mm in target explosive as well as in Teflon were used. The mesh density is enough [10, 37, 38] for Lee-Tarver model to sufficiently resolve the reaction zone. The thickness of the gauges was not mentioned in Vandersall et al., therefore we assume that it would be the same as used in recent experiments carried out in the same laboratory by the authors [39]. The gauges were placed at 0, 8, 14, 20, 25 and 28 mm from the impact surface of the target, and were modelled by Teflon with the Grüneisen equation of state [40], while the target LX-17 was modelled by Lee-Tarver equation of state [10, 37, 38]. The corresponding parameters are given in Table 1 and Table 2 respectively.

| Table 1. Material parameters and Grüneisen equation of state for the Teflon flyer |
|-------------------------------------|-------------------------|----------------|---------------------|----------|----------|----------|----------------|
| Density ($\rho_0$) | Shear modulus ($G$) | Yield Stress ($\sigma$) | Sound Speed ($C_{(cm/\mu s)}$) | Grüneisen equation of state parameters |
| 2.15 g/cm$^3$ | 2.33 Mbar | 0.0050 Mbar | 0.134 cm/µs | $S_1$ | $S_2$ | $S_3$ | $\gamma_0$ | $\alpha$ |

Initial density $\rho_0 = 1.905$ g/cm$^3$

| Table 2. The parameters for modified Lee-Tarver model for LX-17 |
|-------------|-------------------------|-------------------------|-------------------------|-------------------------|
| Reactants JWL EOS | $A = 778.1$ Mbar | $B = -0.05031$ Mbar | $R_1 = 11.3$ | $R_2 = 1.13$ | $\omega = 0.8938$ | $C_v = 2.487 \times 10^{-5}$ Mbar/K | $T_0 = 298^\circ$K |
| Product JWL EOS | $A = 14.8105$ Mbar | $B = 0.6379$ Mbar | $R_1 = 6.2$ | $R_2 = 2.2$ | $\omega = 0.5$ | $C_v = 1.0 \times 10^{-5}$ Mbar/K | $E_0 = 0.064$ Mbar |
| Reaction Rates | $I = 4.0 \times 10^6$ µs$^{-1}$ | $\lambda_{\text{lim}} = 0.02$ | $G_1 = 4500$ Mbar$^2$µs$^{-1}$ | $G_2 = 30$ Mbar$^2$µs$^{-1}$ | $\gamma = 3.0$ | $c = 0.667$ | $g = 0.667$ | $z = 1.0$ |
| Desensitization Parameters | $n = 1$ | $P_0 = 96$ Kbar | $C_f = 0.1$ |

In the present simulation, as a first step, we were able to get the reported impact pressures and the time durations using a 3.5 mm Teflon flyer impact LX-17 target covered by 1.5 mm thick Teflon buffer plate. Later on, simulations were performed using this arrangement for application of the desensitization model for LX-17.
The requirement for our model is the desensitization criteria $P^\tau$ for LX-17, which is not available at present. However we tried different values of $n$ and $P^\tau$ to have as good match to the experimental pressure history records as possible. We find that $n=1$ and $P^\tau=9.6$ gives the best match, and these values are used in the present calculations. It implies that for LX-17, the desensitization rate linearly varies with pressure. This may be the reason why Vandersall et al. was able to reproduce the experimental data with Oliveira’s model. However such results can also be obtained with other pairs of $n$ and $P^\tau$, which reflects the flexibility of the model as well as emphasises the need to perform further experiments to exactly determine the desensitization criterion.

The thickness of the LX-10 booster was adjusted so as to make the detonation wave arrive at gauge point at 28 mm depth at the time as given in the experiment (plot 3 in Vandersall et al.). In case of shot # 4817, the weak shock travels inside the explosive up to the depth of 22.6 mm at time 8.5 µs when it interacts with the detonation wave arriving from opposite direction. It can be observed from the experimental results as well as in calculation results that the pressure at the gauge points the material with increasing degree of desensitization. Within one microsecond it comes across the fully desensitized material with maximum degree of desensitization ($\varphi=1$), and the pressure starts decreasing at the next gauge points (at 14, 8 and 0 mm depths).

In case of original Lee-Tarver model without any desensitization treatment, the calculation results do not reproduce the experimental results, as shown in Figure 2. However using the modified model, the match between the calculated and experimental pressure histories is excellent, as shown in Figure 4. The dotted lines in Figures 2-4 represent the calculated histories while the solid lines represent the
experimental histories. In case of shot # 4829, no desensitization was observed, and the comparison between the experimental and calculated pressure histories is shown in Figure 4.

![Figure 4](image)

Figure 4. The experimental (shot # 4829) and calculated pressure histories at various gauge points inside the target using modified Lee-Tarver model. No desensitization was observed.

3. Conclusions
The present modification of Lee-Tarver model can be used to account for the desensitization effects. The extension does not change the functionality of the original Lee-Tarver model, unless the explosive is subjected to a weak preschok, which desensitizes the explosive. The present study demonstrates that the use of the desensitization criterion \((n=1, P^nτ=9.6)\) for LX-17 yields an excellent match with the experimental results. However, such pair is just estimation; the desensitization criterion \((n, P^nτ)\) is needed to be exactly determined for TATB based explosives by further desensitization experiments such as in references [9, 12] for simulation of multiple compression effects. The desensitization model used here is flexible enough to make use of other \((n, P^nτ)\) pairs by modifying the empirical parameters accordingly.

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5. References
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