Critical deflagration criterion of PTFE/Al/W reactive materials

Yuanfeng Zheng*, Hongbing Ma, Huanguo Guo and Chenghai Su

1 State Key Laboratory of Explosion Science and Technology, Beijing Institute of Technology
E-mail: zhengyf@bit.edu.cn

Abstract: The unique reaction regime of reactive materials includes processes of impact-induced fracture, pre-ignition, post-perforation dispersion, local initiation, and deflagration. The complex interaction between reactive materials and target leads to difficulty in describing critical deflagration criterion of reactive materials. As such, a fundamental projectile/target interaction condition for the complete deflagration of reactive material fragment is derived based on the 1-dimensional shock wave relationships. The critical deflagration pressure $P_c$ of reactive materials is fitted based on the energy release experiments of reactive material fragments. The results show the importance and necessity of the fracture behavior to the deflagration, and the deflagration behavior depends on impact velocity, geometry of reactive material fragment, target material and thickness, et al. As for the deflagration of fractured reactive materials behind the target, the critical deflagration size for reactive material debris is obtained based on the Grady energy model.

1. Introduction
As a typical of non-sustaining reaction reactive materials, the PTFE-based reactive materials have received more and more attention in the last twenty years. The wide applications in military purpose may include reactive fragments in fragmentation warheads, reactive liners in shaped charge warheads, reactive rods in penetration warheads, and so on. Meanwhile, the experiences show that the reactive liners suffering strong shock loadings during formation are likely to react completely. However, in most cases, the reactive fragments and reactive rods cannot react absolutely due to insufficient impact process. For example, if a reactive fragment with a mass of 10g impacts an aluminum plate with a thickness of 1mm, the insufficient interaction between fragment and plate may only lead to a small portion of the reactive fragment occurring deflagration, and most of the reactive fragment only burns, fractures, deforms and even keeps intact. With the change of impact condition, the deflagration mass of reactive fragment varies correspondingly. As such, it is necessary to study the critical deflagration conditions of reactive materials [1-3].

The present paper firstly performs a series of ballistic experiments to reveal the influences of
impact velocity on energy release of reactive materials. The deflagration mechanism is discussed and a critical criterion is proposed to explain the deflagration behavior. At last, the critical pressure and the critical fracture size are obtained.

2. Experimental details

2.1. Reactive material samples
The impact-initiated reactive materials are formed by mixing two kinds of metal powders, such as aluminum (Al) and tungsten (W), into the PTFE matrix in the present paper. The theoretical density of the reactive materials is 2.71g/cm$^3$. The initial powders have an approximate spherical shape with the following average sizes: 100nm PTFE powder, 200μm Al and 80μm W. Actually, PTFE powder is chosen for its excellent formability and acts as the binder of the mixture. Moreover, PTFE decomposes during the penetration process and releases a large amount of fluoride gases, which would be used as the oxidizing agents for the reaction system. W powder is used as an indispensable constituent for its high density. Al powder would react violently with the fluoride gases, releasing much chemical energy and gaseous products [4].

2.2. Reactive material sample preparation
The PTFE/Al/W reactive material samples are prepared, based on the following steps [5-6]:

(1) First, the powders are mixed by a planetary mill machine for 2h.

(2) Second, 10.0g of well-mixed powder is weighed and placed in a pressing mold. A cold pressing pressure of 200MPa is maintained for 10s. The pressed samples are then relaxed at ambient pressure and temperature for 24h to remove the residual stress.

(3) Third, the pressed samples are then sintered in a nitrogen atmosphere with a maximum temperature of 380°C.

Photographs of PTFE/Al/W samples are shown in figure 1.

![Figure 1. Photographs of PTFE/Al/W reactive material samples.](image)

2.3. Setup
The experimental setup is shown in figure 2. The reactive material sample is accelerated by a 25mm caliber ballistic gun. Then, the sample impacts the test chamber with a certain velocity. The test chamber is cylindrical with a volume of 27L. The front of the test chamber is composed of flange, aluminum plate, rubber sealing ring and bolts. The aluminum plate is a replaceable component with a thickness of 6mm. The rear of the test chamber is made up by flange, steel plate, rubber sealing ring and bolts. The thickness of the rear steel plate should be sufficient to ensure that it will not be
perforated by the residual projectile. The thickness of the chamber side should ensure that no obvious deformation occurs during the test. In order to observe the deflagration behavior of reactive materials, a transparent observation window is set on the side of the chamber. Pressure sensor is set on the tank body.

![Experimental setup.](image)

By adjusting the black powder mass, the impact velocities are controlled within the range from 800m/s to 1700m/s. The overpressure inside the test chamber is measured by the pressure sensor.

3. Experimental results
A series of ballistic experiments are conducted. The typical high speed camera photographs are presented in figure 3, and one representative overpressure curve is shown in figure 4. All the experimental data is listed in table 1. The results show that the measured overpressure inside the chamber depends on the impact velocity strongly. For the mechanism considerations, the impact between the reactive material sample and the aluminum plate may result in different response behavior, including deflagration, burn, fracture, and deformation. The response behavior is decided by the shock intensity caused by the impact process. Actually, figure 5 clarifies this complex response process. If the sample impacts the aluminum plate with a mild velocity, the pressure inside the sample would decrease gradually along the axis. At the impact interface, the pressure is high enough and results in the reactive materials near the impact interface react in the form of deflagration. Unlike the traditional explosives, the reactive materials discussed here cannot occur self-sustaining reaction. That is to say, the deflagration reaction needs the shock wave high enough to activate the reactive materials continuously. However, the shock wave inevitably decreases with the propagation process. As such, the reactive materials far from the impact interface may only react in the form of combustion. With the shock wave further decreases, the reactive materials near the rear of the sample may only fracture, deform, or even keep intact. Meanwhile, it should be stressed that only that portion occurring deflagration reaction has a contribution to the overpressure, which indicates that the value of overpressure represents the mass of deflagrated reactive materials. Based on the discussion above, a conclusion can be drawn that there is a critical pressure ($P_c$) for the deflagration of reactive materials, which can be obtained approximately based on the data listed in table 1.
4. Critical deflagration pressure

In fact, the condition for the whole sample deflagrating can be summed up as follows: the initial shock wave formed in the process of impact reaches the bottom of the sample before the reflection wave catching up, and the initial shock wave should high enough to ensure the pressure at the sample rear being higher than the critical deflagration pressure $P_c$. 

**Table 1.** Experimental data.

| No. | Impact velocity (m/s) | Impact pressure (GPa) | Overpressure inside (MPa) | No. | Impact velocity (m/s) | Impact pressure (GPa) | Overpressure inside (MPa) |
|-----|-----------------------|-----------------------|---------------------------|-----|-----------------------|-----------------------|---------------------------|
| 1   | 833                   | 4.29                  | 0.29                      | 7   | 1359                  | 8.04                  | 0.50                      |
| 2   | 927                   | 4.91                  | 0.30                      | 8   | 1407                  | 8.42                  | 0.54                      |
| 3   | 1035                  | 5.64                  | 0.32                      | 9   | 1450                  | 8.76                  | 0.58                      |
| 4   | 1193                  | 6.78                  | 0.38                      | 10  | 1551                  | 9.58                  | 0.62                      |
| 5   | 1253                  | 7.23                  | 0.40                      | 11  | 1602                  | 10.0                  | 0.59                      |
| 6   | 1301                  | 7.59                  | 0.46                      | 12  | 1643                  | 10.4                  | 0.61                      |

**Figure 3.** Typical high speed camera photographs of reactive material sample releasing energy.

**Figure 4.** Representative overpressure curve. **Figure 5.** Response behavior during impact.
Firstly, the condition for the shock wave sweeping the whole sample is studied. Based on the conservation of mass, momentum and energy,

\[ \rho_0 U = \rho (U - u) \]  \hspace{1cm} (1) 

\[ P - P_0 = \rho U u \]  \hspace{1cm} (2) 

\[ E - E_0 = (P + P_0)(V_0 - V) / 2 \]  \hspace{1cm} (3) 

where, \( \rho_0, E_0, V_0, P_0 \) are initial density, internal energy, specific volume and pressure. \( \rho, E, V, P \) are density, internal energy, specific volume and pressure after shock. \( U \) and \( u \) are shock wave velocity and particle velocity, respectively.

The relationship between \( U \) and \( u \) is

\[ U = c_0 + su \]  \hspace{1cm} (4) 

where, \( c_0 \) is material sound velocity, and \( s \) is a constant.

Due to the continuity condition on the impact interface,

\[ v_i = u_p + u_t \]  \hspace{1cm} (5) 

\[ P_p = P_t \]  \hspace{1cm} (6) 

where, \( v_i \) is impact velocity; \( u_p \) and \( u_t \) are particle velocities in reactive material sample and in target, respectively; \( P_p \) and \( P_t \) are shock wave pressure in reactive material sample and in target, respectively.

Substituting equation (4) into equation (2),

\[ P = \rho_0 (c_0 + su) u \]  \hspace{1cm} (7) 

The pressures inside the reactive material sample and the target can be expressed as,

\[ P_p = \rho_{0p} (c_0 + s_p (v_i - u_i))(v_i - u_i) \]  \hspace{1cm} (8) 

\[ P_t = \rho_{0t} (c_0 + s_t u_t) u_t \]  \hspace{1cm} (9) 

Substituting equations (8) and (9) into equation (6),

\[ \rho_{0p} (c_{0p} + s_p (v_i - u_i))(v_i - u_i) = \rho_{0t} (c_{0t} + s_t u_t) u_t \]  \hspace{1cm} (10) 

Thus, the particle velocity in target can be obtained as

\[ u_t = [-b \pm (b^2 - 4ac)^{0.5}] / (2a) \]  \hspace{1cm} (11) 

where
\[ a = \rho_{0p} s_p - \rho_{0i} s_i \]
\[ b = -(2 \rho_{0p} s_p v_i) - (\rho_{0p} c_{0p}) - (\rho_{0i} c_{0i}) \]
\[ c = (\rho_{0p} v_i c_{0p}) + (\rho_{0p} v_i^2 s_p) \]

And

\[ U_i = c_{0i} + s_i u_i \quad (12) \]

Substituting \( u_i \) and \( U_i \) into equation (1), the target density after shock should be

\[ \rho_i = \frac{\rho_{0i} U_i}{U_i - u_i} \quad (13) \]

The equation (5) can also be written as

\[ u_p = v_i - u_i \quad (14) \]

And

\[ U_p = c_{0p} + s_p u_p \quad (15) \]

Thus, the density of reactive material sample after shock should be

\[ \rho_p = \frac{\rho_{0p} U_p}{U_p - u_p} \quad (16) \]

Care has to be taken that the shock wave propagates forward into the target and backward into the reactive material sample at the same time. When the shock wave is transmitted to the back of the target plate, a reflection-induced rarefaction wave begins to propagate reversely. If the shock wave propagating inside the reactive material sample is caught up by the rarefaction wave, the shock wave pressure in the sample would drop rapidly, which may terminate the fracture and reaction behavior. In hence, the minimum thickness of the target plate should meet the requirement that when the shock wave sweeps through the whole reactive material sample, the rarefaction wave just arrives at the rear of the sample. The time required for the shock wave to sweep through the sample can be expressed as

\[ t_{0p} = t_{0i} + t_i + t_p \quad (17) \]

where

\[ t_{0p} = \frac{L_p}{U_p} \quad t_{0i} = \frac{L_i}{U_i} \quad t_i = \frac{L_i \rho_{0i}}{\rho_i C_i} \quad t_p = \frac{L_p \rho_{0p}}{\rho_p C_p} \]
$t_{op}$ and $t_{ot}$ are times for the shock wave sweeps the reactive material sample and the target, respectively. $t_p$ and $t_t$ are times for the rarefaction wave sweeps the reactive material sample and the target, respectively. $L_t$ is the target thickness; $L_p$ is the sample length; $U_p$ and $U_t$ are shock wave velocities in the reactive material sample and in the target, respectively; $C_p$ and $C_t$ are rarefaction wave velocities in the reactive material sample and in the target, respectively. $\rho_p$ and $\rho_t$ are densities of reactive material sample and the target, respectively.

The rarefaction wave velocity can be estimated as

$$C = U \left\{ 0.49 + \left[ \frac{(U - u_t)}{U} \right]^2 \right\}^{0.5}$$

(18)

Thus, the minimum target plate thickness can be obtained by combining equations (17) and (18),

$$L_{t_{min}} = L_p \frac{\left( \frac{1}{U_p} - \frac{\rho_{op}}{\rho_{p}} \right) - \frac{\rho_{op}}{\rho_{p} C_p}}{\frac{\rho_{ot}}{\rho_t C_t} + 1/U_t}$$

(19)

The second, but equally important, is the pressure of the shock wave. The initial shock wave pressure should high enough to ensure the pressure at the end of the sample being higher than the critical deflagration pressure $P_c$. Thus

$$P(L_p) = P_p \exp(-\alpha L_p) \geq P_c$$

(20)

where, $P(L_p)$, $P_p$ and $\alpha$ are shock wave pressure at the end of the sample, initial shock wave pressure, and shock attenuation coefficient in reactive materials.

Finally, the condition for the whole sample deflagrating is

$$L_t \geq L_p \frac{\left( \frac{1}{U_p} - \frac{\rho_{op}}{\rho_{p}} \right) - \frac{\rho_{op}}{\rho_{p} C_p}}{\frac{\rho_{ot}}{\rho_t C_t} + 1/U_t}$$

$$P(L_p) = P_p \exp(-\alpha L_p) \geq P_c$$

(21)

The Hugoniot parameters of reactive materials can be estimated from reference [7]. Based on equation (19), the relationship between minimum aluminum plate thickness and impact velocity can be drawn in figure 6. The calculated results indicate the aluminum plates used in the experiments are thick enough to ensure the shock wave propagating to the end of the sample.
Figure 6. The relationship between minimum aluminum plate thickness and impact velocity.

In order to get the value of parameter $P_c$, a variable, named deflagration ratio, is introduced and defined as

$$\eta = \frac{m}{M_h}$$  \hspace{1cm} (22)

where, $m$ and $M_h$ are deflagration mass and the total mass of the sample, respectively.

Based on the previous study [8]

$$\eta = \frac{m}{M_h} = \frac{mq}{M_h q} = \frac{Q}{Q_{\text{max}}} = \frac{\Delta P}{\Delta P_{\text{max}}}$$  \hspace{1cm} (23)

where, $q$ is the heat output per gram of reactive materials, and $\Delta P$ is the pressure peak inside the test chamber. The subscript “max” means the whole sample reacting in the form of deflagration.

Finally, based on the equation (23) and the experimental data in section 3, the deflagration ratio can be obtained and listed in table 2.

| No. | Impact velocity (m/s) | Impact pressure (GPa) | Deflagration ratio (%) | No. | Impact velocity (m/s) | Impact pressure (GPa) | Deflagration ratio (%) |
|-----|-----------------------|-----------------------|------------------------|-----|-----------------------|-----------------------|------------------------|
| 1   | 833                   | 4.29                  | 46.8                   | 7   | 1359                  | 8.04                  | 80.6                   |
| 2   | 927                   | 4.91                  | 48.4                   | 8   | 1407                  | 8.42                  | 87.1                   |
| 3   | 1035                  | 5.64                  | 51.6                   | 9   | 1450                  | 8.76                  | 93.5                   |
| 4   | 1193                  | 6.78                  | 61.3                   | 10  | 1551                  | 9.58                  | 100                    |
| 5   | 1253                  | 7.23                  | 64.5                   | 11  | 1602                  | 10.0                  | 95.2                   |
| 6   | 1301                  | 7.59                  | 74.2                   | 12  | 1643                  | 10.4                  | 98.4                   |

As for a cylindrical sample, the deflagration ratio can be revised as
\[ \eta = \frac{m}{M_n} = \frac{x}{l} \]  

(24)

where, \( l \) is the total length of the reactive material sample.

Meanwhile, the relationship between \( P_0 \) and \( P_c \) is

\[ P_c = P_0 \exp(-\alpha \alpha) \]  

(25)

Combining equations (25) and (26),

\[ \eta = -\frac{1}{\alpha d} \ln \frac{P_c}{P_0} \]  

(26)

The equation (27) can also be written as

\[ \eta = -\frac{1}{\alpha d} \ln \frac{P_c}{P_0} = -\frac{1}{\alpha d} (\ln P_c - \ln P_0) = -\frac{1}{\alpha d} \ln P_c + \frac{1}{\alpha d} \ln P_0 = C_1 + C_2 \ln P_0 \]  

(27)

where, \( C_1 \) and \( C_2 \) are constants. \( C_1 = -\frac{1}{\alpha d} \ln P_0, \quad C_2 = \frac{1}{\alpha d} \).

Based on the data in table 2, equation (27) is fitted and the corresponding curve is shown in figure 7. Parameters \( P_c \) is fitted as 2.46GPa.

5. Critical average fracture size

For mechanism considerations, the fracture behavior of reactive materials is of importance to the deflagration. In fact, the sample fractures and forms a reactive material debris in the test chamber, and it is the reactive material debris that occurs deflagration reaction inside. Herein, the average fracture size of the debris is introduced to characterize the deflagration ratio. With the impact velocity increases, the average fracture size decreases, which would improve the deflagration ratio.

If all the debris is assumed to be spherical,
\[ d = \left( \frac{1.2Y}{\rho(d\varepsilon/dt)^2} \right)^{1/2} \]  

(28)

where, \( Y \) and \( \rho \) are yield strength (about 20.5MPa) and material density, respectively. \( d\varepsilon/dt \) is the strain rate of reactive material sample, which can be estimated by simulations [9].

As discussed above, when the impact velocity is 1551m/s, the deflagration ratio reaches about 100%. As such, the average fracture size in this case is considered as the critical average fracture size \( d_c = 0.91 \) mm.

6. Conclusions

Critical deflagration criterion of PTFE/Al/W reactive materials is studied in the present paper, including critical pressure and critical average fracture size. Ballistic gun experiments of reactive material sample against sealed test chamber are conducted and the results show the overpressure inside the chamber is highly influenced by the impact velocity, indicating a non-sustaining reaction behavior of the reactive materials. There are two conditions for a reactive material sample deflagrating with a deflagration ratio of 100%. On one hand, the initial shock wave formed during impact needs to reach the bottom of the sample before the reflection wave catching up; on the other hand, the initial shock wave should high enough to ensure the pressure at the sample rear being higher than the critical deflagration pressure. By fitting experimental data, the critical deflagration pressure and the critical average fracture size are obtained as 2.46GPa and 0.91mm, respectively.

Reference

[1] DE Technologies 2006 Inc. Reactive Fragment Warhead for Enhanced Neutralization of Mortar Rocket & Missile Threats ONR-SBIR: N04-903 http://www.detk.com
[2] J Nable, A Mercado and A Sherman 2006 Novel Energetic Composite Materials Proc. Symp. Mater. Res. Soc. 896 0896-H01-03
[3] E L Baker, A S Daniels, K W Ng, V O Martin and J P Orosz 2001 19rd International Symposium on Ballistics Interlaken Switzerland May 7-11 p 569-71
[4] Yuanfeng Zheng, Chenghai Su, Huanguo Guo, Qingbo Yu and Haifu Wang 2019 Jet. Propellants, Explosives, Pyrotechnics 44 1-11
[5] Huanguo Guo, Jianwan Xie, Haifu Wang, Qingbo Yu and Yuanfeng Zheng 2019 Penetration Behavior of High-Density Reactive Material Liner Shaped Charge Materials 12 3486
[6] Li Y, Jiang C L, Z C and Niu H H 2017 Materials 10 175
[7] Batsanov S S 1994 Effects of explosions and materials New York: Springer-Verlag
[8] Haifu Wang, Yuanfeng Zheng, Qingbo Yu, Zongwei Liu and Weimin Yu Impact-induced initiation and energy release behaviour of reactive materials JOURALA OF APPLIED PHYSICS 110 074904(2011).
[9] M Kipp, D Grady and J Swegle 1993 Int. J. Impact. Eng. 14 427-38