LLNL-TR-681467

LAWRENCE LIVERMORE NATIONAL LABORATORY

ONE-DIMENSIONAL TIME TO EXPLOSION (THERMAL SENSITIVITY) TESTS ON PETN, PBX-9407, LX-10, AND LX-17

P. C. Hsu, S. Strout, M. McClelland, F. Ellsworth

January 28, 2016

This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

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Peter. C. Hsu, Steve Strout, Matthew McClelland, and Fred Ellsworth

December 8, 2015

Lawrence Livermore National Laboratory, Livermore, CA USA

Abstract. Incidents caused by fire and combat operations can heat energetic materials that may lead to thermal explosion and result in structural damage and casualty. Some explosives may thermally explode at fairly low temperatures (< 100 C) and the violence from thermal explosion may cause a significant damage. Thus it is important to understand the response of energetic materials to thermal insults. The One Dimensional Time to Explosion (ODTX) system at the Lawrence Livermore National Laboratory has been used for decades to measure times to thermal explosion, threshold thermal explosion temperature, and determine the kinetic parameters of thermal decomposition of energetic materials. Samples of different configurations (pressed part, powder, paste, and liquid) can be tested in the system. The ODTX testing can also provide useful data for assessing the thermal explosion violence of energetic materials. This report summarizes the results of our recent ODTX experiments on PETN powder, PBX-9407 pressed part, LX-10 pressed part, LX-17 pressed part and compares the test data that were obtained decades ago with the older version of ODTX system. Test results show the thermal sensitivity of various materials tested in the following order: PETN> PBX-9407 > LX-10 > LX-17.

1. Introduction

Accidents involving with thermal explosion (or cook-off) of energetic materials are costly. Over the last few decades, there has been considerable research effort on the thermal decomposition and thermal explosion violence of energetic materials at elevated temperatures in different sample geometries and confinement [1-3]. Thermal explosion studies on various energetic materials in two-dimensional geometry such as the Scaled-Thermal-Explosion-Experiment (STEX) system [4] and the Sandia-Instrumented-Thermal-Ignition (SITI) system have been reported [5]. The One Dimensional Time to Explosion (ODTX) system at the Lawrence Livermore National Laboratory (LLNL) has been used since 1970s for cook-off study [6-10]. It is attractive because of the one-dimensional geometry, a minimal sample requirement (up to 2 grams for each test) and low cost. ODTX testing generates three useful technical data: (1) lowest temperature at which thermal explosion would occur (threshold temperature, T_{li}); (2) times to thermal explosion at temperatures above T_{li} for the calculation of activation energy and frequency factor; and (3) thermal explosion violence.

2. A brief description of ODTX capability at LLNL

Understanding the response of energetic materials to thermal incidents is very important for the handling, storage and transportation of energetic materials. The uniqueness of the ODTX system at LLNL is because of its capability to generate three important technical data, which are described below.

(1) lowest temperature at which thermal explosion would occur (T_{li}),
(2) times to thermal explosion at temperatures above T_{li} for the calculation of activation energy and frequency factor for thermal decomposition kinetics,
(3) Thermal ignition/explosion violence

(1) $\boldsymbol{T_{li}}$

Knowing the lowest thermal explosion temperature (T_{li}) for each energetic material is very important for safe storage and transportation to avoid incidental detonation. Two possible scenarios for causing incidental thermal explosions are described below:

1. Energetic materials may be kept and stored in closed containers that are exposed to hot climates. During the summer in some desert areas, outdoor temperature may exceed 120°F while the surface temperature of metallic storage containers exposed to the sun may exceed 170°F (77°C). Given enough time, some energetic materials may ignite and explode.
2. If containers storing energetic materials are kept inside a parked van or truck with windows closed for an extended period of time, the air inside the van may exceed 170°F in the summer. Given enough time, some energetic materials in the containers may ignite and explode.

(2) ***Time to Explosion Data, Activation Energy, and Frequency Factor***
Times to thermal explosion at temperatures above T_{li} for the calculation of activation energy and frequency factor as well as the decomposition kinetics parameters represented by a single-step Prout-Tompkins (Arrhenius) model (shown in the modeling section).

(3) ***Thermal Explosion Violence***
Violence from thermal explosion is an important parameter for cook off study. After the ODTX testing, each anvil was scanned with a surface profilometer to determine the cavity volume increase. Figure 1 shows anvils before and after the thermal explosion from the ODTX testing. The violent thermal explosion discolored the anvils and created craters in the anvils.

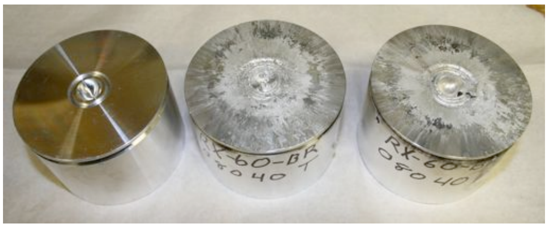


Figure 1. Anvils before and after thermal explosion of a liquid explosive; left was the pristine anvil; also shown are top anvil (middle) and bottom anvil (right) after the thermal explosion.

3. Comparison of ODTX with DSC

Both ODTX and DSC can generate thermal kinetic data for homogeneous explosive samples. For heterogeneous sample mixtures, ODTX is preferred because its sample size of 2 grams is much larger than the DSC sample size of 0.3 mg. Below are several things that ODTX can generate and DSC cannot.

1. The minimum temperature for thermal explosion to occur

   Knowing the minimum temperature for thermal explosion to occur is very important for safe storage and handling of energetic materials. The ability of testing explosive in the ODTX system at lower temperatures until no thermal explosion occurs allows for the determination of fairly precise threshold temperature for thermal explosion to occur. Both DSC and Cheetah do not provide the information.

2. Thermal explosion violence

Violence from thermal explosion is one of the important parameters for explosive safety. Careful monitoring of anvil cavity volume increase before and after thermal explosion allows for the determination of thermal explosion violence. DSC and Cheetah cannot predict nor measure the thermal explosion violence.

3. Samples of different density and configuration
   Samples of any density and any configurations (solid pressed parts, powders, pastes, and liquids) can be tested in the ODTX system. Sample size is much larger than that for DSC (2.0 g for ODTX and ~ 0.3 mg for DSC). Since DSC sample size is less than 1.0 mg, it is difficult to get a good representative sample for a heterogeneous mixture. Thus obtaining thermal decomposition kinetic on heterogeneous mixtures from DSC data may not be accurate.

4. System Description and Experiments

The ODTX system, as shown in figure 2, is operated remotely in a test cell. The testing involves heating a 1.27-cm diameter spherical sample in a spherical cavity between two aluminum anvils. The sample is remotely delivered to the anvil cavity via the sample delivery system when the anvils reach a predetermined temperature. A microphone sensor measures a sound signal, which indicates the time at which a thermal explosion occurs. The detail description of the LLNL ODTX system can be found elsewhere [11].

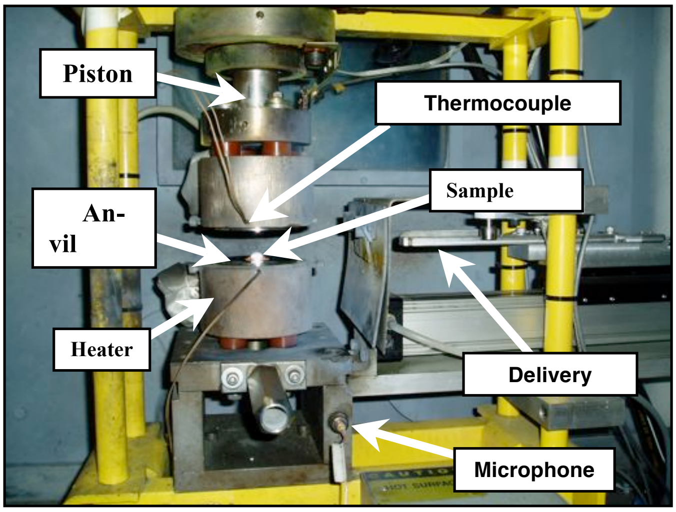


Figure 2. LLNL ODTX system (in service since FY2001).

Samples of various configurations (pressed parts, cast parts, powders, pastes, and liquids) can be tested in the ODTX system. Pressed and cast samples are loaded into the cavity of aluminum anvils directly without secondary containment. An aluminum shell is used as a secondary containment to hold powder samples, pasty samples, or liquid samples before loading to the system. ODTX tests are typically run 10 to 20 times at various temperatures to obtain time to thermal explosion charts. The tests require a total of 30 grams of material.

5. ODTX test results

5.1 ODTX test data on PETN powder, PBX-9407, LX-10, and LX-17

We used the ODTX system to characterize the thermal safety of PETN powder, PBX-9407 pressed part, LX-10 pressed part, and LX-17 pressed part. Totally, 22 ODTX shots were performed from March 2015 to June 2015. 0.94 Grams of PETN powder was packed into a 0.5-inch aluminum spherical shell for ODTX testing. The tests (9 shots) were performed over a range of temperature, from 120 C to 201 C and the times to thermal explosion are shown in Table 1. This is the first time that the thermal sensitivity of PETN in powder form has even been evaluated in the ODTX system. The remainder of 13 ODTX shots was done on pressed parts without the use of aluminum shell for PBX-9407 (4 shots), LX-10 (5 shots), and LX-17 (4 shots), respectively, see Figure 3 for sample pictures. The primary goal of these 13 ODTX shots was to verify and confirm the tests that were performed prior to FY2001 in an older version of ODTX system. Times to thermal explosion are shown in Tables 2, 3, and 4, respectively. The data are plotted in Figure 4. It also shows ODTX data for RDX, HMX, and TATB. The ODTX test results indicated that the thermal sensitivity of PETN powder is similar to that of PETN pressed parts. PETN is more sensitive to thermal insults than HMX, RDX, and TATB and their formulations. The ODTX data obtained from these 22 shots verify the consistency of the ODTX system performance, when compared with data collected in an older ODTX system prior to FY2001.

PETN PBX-9407 LX-10 LX-17

Figure 3. Pictures of spherical samples (0.5” diameter): PETN powder (0.9378 g packed inside a spherical aluminum shell, 50% TMD), PBX-9407, LX-10, and LX-17.

Table 1. ODTX Data for PETN powder (0.94 g or 50% TMD)

Sample ID	**Date tested**	**Temperature, C**	**Times to explosion, seconds**
1	03/30/15	161.9	261
2	03/31/15	149.2	854
3	04/01/15	181.0	76.0
4	04/07/15	201.2	27.3
5	04/08/15	145.8	1,211

6	04/09/15	139.0	3,639
7	04/10/15	132.4	9,267
8	04/14/15	191.9	48.9
9	06/24/15	120	44,568

Table 2. ODTX Data for PBX-9407 pressed part (1.913 g)

Sample ID	**Date tested**	**Temperature, C**	**Times to explosion, seconds**
1	04/15/15	220.4	51.2
2	04/16/15	235.5	7.3
3	04/17/15	193.6	1,721
4	04/21/15	204.2	286

Table 3. ODTX Data for LX-10 pressed part (2.034 g)

Sample ID	**Date tested**	**Temperature, C**	**Times to explosion, seconds**
1	04/22/15	205.2	2,596
2	04/28/15	259.5	152.3
3	04/29/15	224.6	552.4
4	04/30/15	195.1	10.536
5	05/05/15	275.1	39.0

Table 4. ODTX Data for LX-17 pressed part (2.071 g)

Sample ID	**Date tested**	**Temperature, C**	**Times to explosion, seconds**
1	05/13/15	267.6	3,782
2	05/14/15	293.1	1,045
3	05/15/15	321.7	223.5
4	05/28/15	253.2	8,866

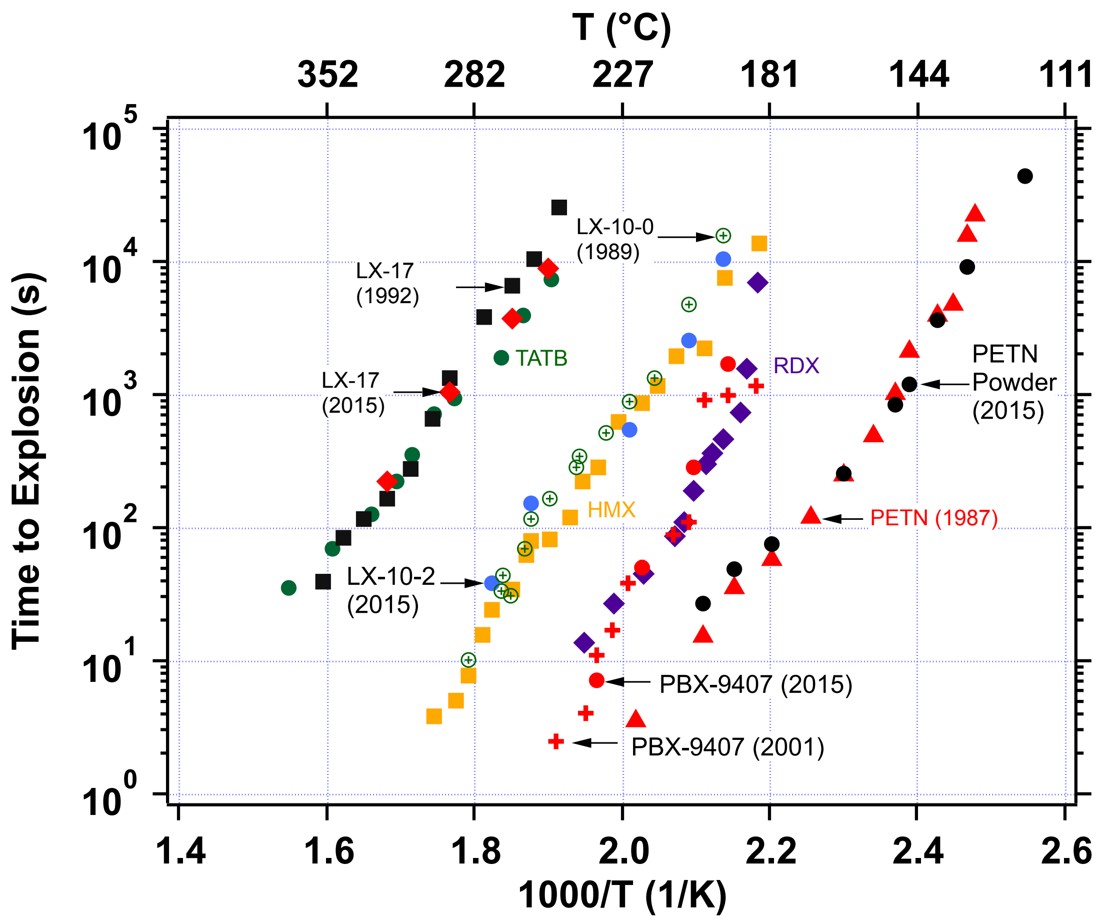


Figure 4. ODTX test data of PETN powder, PBX-9407, LX-10, LX-17 and several commonly used high explosives (all in pressed parts) HMX, PETN, RDX, and TATB.

5.2 Thermal explosion violence (ODTX anvil scanning)

Figure 5 shows the anvils before and after the thermal explosion of PETN powder. The anvils indicated some melting from the extremely hot gas generated by thermal explosion. A new surface profilometer was used to scan the cavity volumes of some spent anvils, see the image in Figure 5. Table 5 lists the cavity volume increases of the aluminum anvils for PETN powder at different temperatures.

Average cavity volume increase for PETN powder is 1.00 cc/g that is much lower than that for PBX-9501 and LX-10. Figure 5 and Figure 6 show the spent anvils and cavity volume expansion. HMX-based formulations (LX-04, LX-07, LX-10, PBX-9501) have very high thermal explosion violence and TATB-based formulations (LX-17, and PBX-9502) have very low thermal explosion violence. In general, the lower the thermal explosion violence is, the better the energetic material is from the thermal safety standpoint.

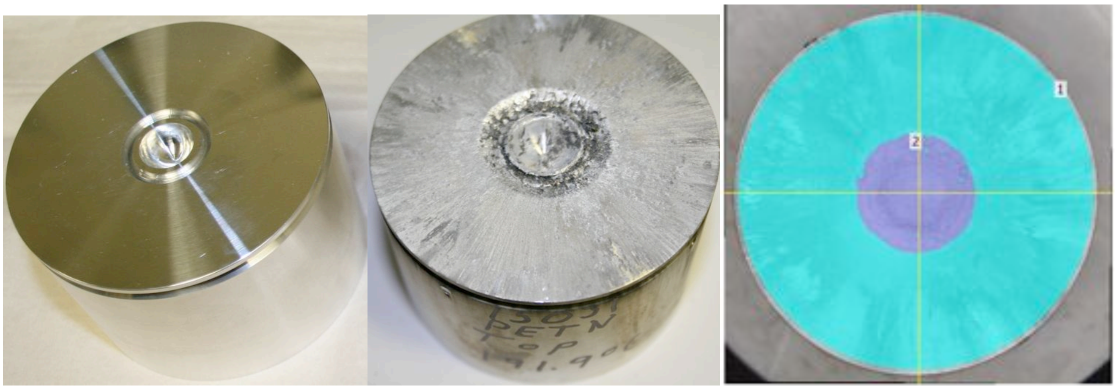
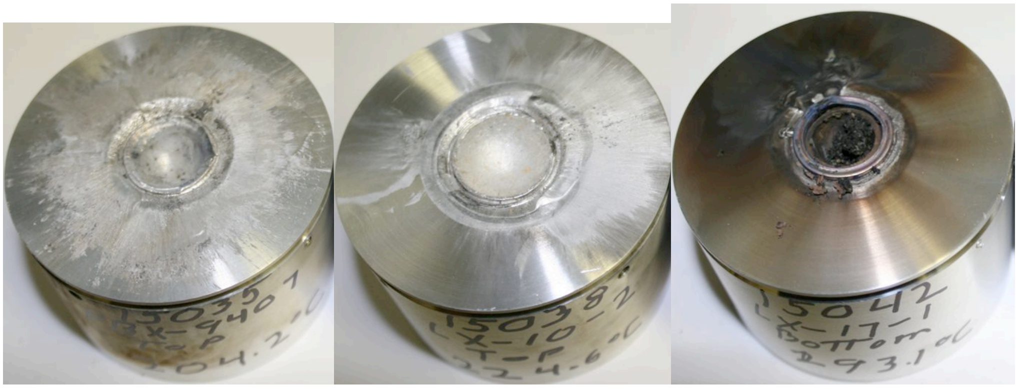


Figure 5. Anvils before and after thermal explosion of PETN powder; top left was the pristine anvil; top middle was spent anvil, top right shows the image of the spent anvil.



PBX-9407 LX-10 LX-17

Figure 6. Spent anvils: PBX-9407 (left), LX-10 (middle), and LX-17 (right); LX-10 shows more violence than PETN, PBX-9407 and LX-17 upon thermal explosion.

Table 5. Cavity volume increases of the spend anvils from ODTX testing on PETN powder

Run ID	Test temperature, C	Cavity volume increases, cc/g
15027	201.2	1.12
15031	191.9	0.88
15026	181.0	0.43
15024	161.9	2.14
15025	149.2	0.62
15028	145.8	1.99
15029	139.0	0.61
15030	132.4	0.60
15047	120	0.62

Average volume increase: 1.00 cc/g.

Table 6. Crater volume increases and relative degree of thermal explosion violence

Material	Average volume increase, cc/g	Relative degree of violence
PBX 9501, pressed	2.57	1.00
PETN powder	1.00	0.39
HPP, casted	0.68	0.26
UN/Al, powder	0.62	0.24
LMI, liquid	0.51	0.20
Semtex 1H, paste	0.19	0.07
Tritonal, casted	0.10	0.04
TATB, pressed	0.07	0.03

Table 7 shows the lowest temperatures at which thermal explosion (threshold temperature, T_{li}) would occur. PETN shows the lowest threshold temperature for thermal explosion among commonly-used high explosives.

Table 7. Threshold temperature for thermal explosion (T_{li}) for several neat explosives

Materials	T_{li}, °C
Home-made explosive liquids	80 to 110
PETN	<120.0
RDX	175.0
HMX	180.0
TNT	200
TATB	230

References

[1] Catalano E, McGuire R, Lee E,Wrenn D, Ornellas D and Walton J 1976 The Thermal Decomposition and Reaction of Confined Explosives”, 6th International Detonation Symposium.

[2] Chidester S K, Tarver C M, Green L G and Urtiew P A 1997 On the Violence of Thermal Explosion in Solid Explosives *Combustion and Flame* **110** *264-280.*

[3] Tarver C M and Tri T 2004 Thermal Decomposition Models for HMX-based plastic bonded explosives *Combustion and Flame* **137** 50-62.

[4] Wardell J F and Maienschein J L 2002 The Scaled Thermal Explosion Experiment 12th International Detonation Symposium, pp 384-393, San Diego August.

[5] Dodd A B and Kaneshige M J 2006 Cook-off Model Development and Analysis of Energetic Materials Using Instrumented Thermal Ignition (SITI) Experimental Data 13th International Detonation Symposium, pp 516-526, Norfolk July.

[6] Williams M R and Matei M V 2006 The Decomposition of Some RDX and HMX Based Materials in the One-Dimensional Time to Explosion Apparatus Part 1 Time to Explosion and apparent Activation Energy *Propellats Explosive Pyrotechnics* **31** 435-441.

[7] Koerner J, Maienschein, J L, Burnham A and Wemhoff A 2007 ODTX Measurements and Simulations on Ultra Fine TATB and PBX 9502, UCRL-Conf-232590, LLNL.

[8] Maienschein J L, Garza R G, Nichols A L and Chandler J B 1996 Kinetics of Thermal Ignition of AP/Al/HTPB Propellants, JANNAF Meeting, Monterey.

[9] Hsu P C, Hust G, Howard M, Chidester S K, Springer, H K and Maienschein J L 2010 The ODTX System for Thermal Ignition and Thermal Safety Sudy of Energetic Materials, 14th International Detonation Symposium, Coeur d’A;eme, Idaho, April 11-16.

[10] Hsu P C, Hust G, Howard M, Chidester SK, Springer H K and Maienschein J L 2011 Study of thermal Thermal Sensitivity and Thermal Explosion Violence of Energetic Material s in the LLNL ODTX System, APS Conference, Chicago, July.

[11] Hsu P C 2009 LLNL OD*T*X System for Thermal Safety Determination of Energetic Materials, LLNL-BR-411732.

[12] Owens & Souers, HE Reference Guide, 2013.

[13] Bastea et al. Cheetah, 2010 edition.