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Insensitive High Explosive Qualification of LLM-105 Based Explosive

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List of Acronyms

CHE	Conventional High Explosive
DAPO	2,6-Diaminopyrazine-1-oxide
DDT	Deflagration-to-Detonation Transition
DOE	Department of Energy
ESC	Explosives Safety Committee
HE	High Explosive
HMX	High Melting Explosive
IHE	Insensitive High Explosive
IHC	Interim Hazardous Classification
LANL	Los Alamos National Laboratory
LLM	Lawrence Livermore Material
OFF	Outside Firing Facility
SDT	Shock-to-Detonation Transition
STD	Standard
TATB	triaminotrinitrobenzene

Abstract

The U.S. Department of Energy (DOE) Explosive Safety Committee (ESC) updated the requirements in DOE-STD-1212-2019 to qualify a material as an Insensitive High Explosive (IHE) in May 2021. The IHE Material and IHE Subassembly Qualification Test Description and Criteria require four tests to qualify a material as an IHE. These test are: 1) Bullet Impact Testing; 2) Shock to Detonation Transition Testing (SDT); 3) Deflagration to Detonation Transition Testing (DDT); and 4) Skid Testing. For the first time since TATB and its formulations were qualified as IHEs under the DOE IHE standard dating from the 1980s, an energetic material is going to undergo testing to qualify as an IHE. An LLM-105-based explosive, formulated with an elastomeric polymer, exhibits IHE behavior in its resistance to DDT, SDT, reaction upon bullet impact, drop impact, and friction. Lawrence Livermore National Laboratory (LLNL) is currently performing a subset of tests required to qualify a material as an IHE with either less replicates than required or on a different scale than that prescribed in the IHE standard. This presentation will discuss the testing performed at the smaller scale and the scope of the full-scale suite of tests required per DOE-STD-1212-2019.

Introduction

For the first time since the 1980's, an explosive is being tested against the newly revised Department of Energy (DOE) Explosive Safety Standard, DOE-STD-1212-2019, Insensitive High Explosive (IHE) Qualification requirements. LLM-105 formulated with an elastomeric polymer is being evaluated for booster application due to its desired performance coupled with known safety traits. The remainder of this report will give an overview of what LLM-105 is, the IHE tests required per DOE-STD-1212-2019 Section 16, and how LLM-105 formulated material performed in each of the tests prescribed in those tests.

LLM-105 Material Overview

Since first synthesized in 1995 at the Lawrence Livermore National Laboratory (LLNL), 2,6-diamino-3,5-dinitropyrazine-1-oxide, also known as LLM-105, has been considered a contender for various insensitive high explosive applications [1]. LLM-105 is thermally stable energetic with a melting point of greater than 370ºC [1] . It has a density of 1.918 g/cm3 [1]. LLM-105 also displays insensitivity to shock, drop impact, friction, bullet impact, and transition from deflagration to detonation. The sensitivity of LLM-105 is between that of HMX and TATB [2]. In addition, it has an energy content that is about 85% of HMX and 15% more than TATB [2].

There have been several routes to synthesizing LLM-105 published. The route of choice is to nitrate 2,6-diaminopyrazine-1-oxide (DAPO) with a mixture of nitric acid and sulfuric acid. This is followed by an ammonium nitrate quench for purification [3]. This gives a 1 step synthesis method that yields high purity LLM-105 [1]. Figure 1 below shows the 1 step DAPO synthesis route to yield LLM-105.

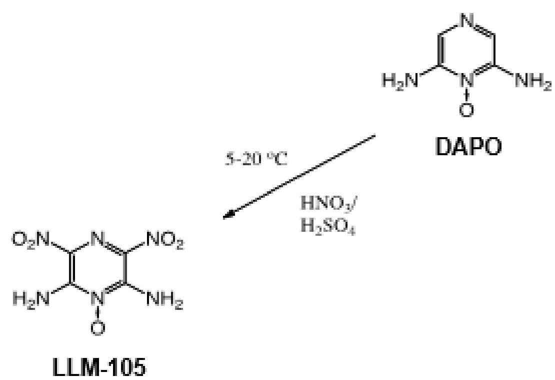

Figure 1. Synthesis of DAPO-LLM-105

The DAPO route is used to synthesize the source LLM-105 material for its formulations with elastomeric polymer, and have been researched over the past 20 years. Figure 2 shows molding powder formulated from LLM-105 with elastomeric binder.

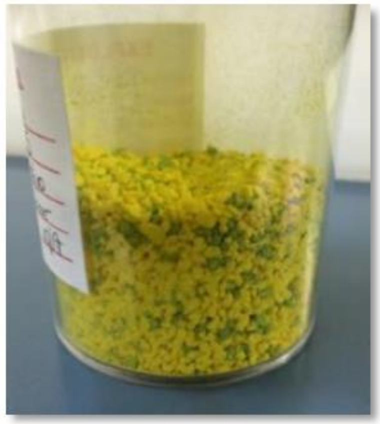

Figure 2. LLM-105 formulated with elastomeric polymer

IHE Qualification Requirements

Per DOE-STD-1212-2019, for a material to be designated an IHE it must demonstrate the following in scales that are conservative for the relevant nuclear weapon application:

- Does not transition from deflagration to detonation (DDT),
- Does not transition from shock to detonation (SDT) under the following three conditions
  - 3.5 GPa, 3 μs 1-dimensional shock insult at 25°C.
  - 5.3 GPa, 0.5 μs 1-dimensional shock insult at 25°C.
  - 1.5 GPa, 3 μs 1-dimensional shock insult when heated to 10°C below the cook-off temperature of the explosive.
- Passes the skid test and bullet test as defined in LLNL-TR-679331-REV-1/LA-UR-15-29238, "IHE Material and IHE Subassembly Qualification Test Description and Criteria." [4]

Additionally, prior to conducting the IHE material tests the material must have an interim hazard classification (IHC) as assigned by the Department of Transportation (DOT), per DOE-STD-1212-2019.

LLM-105 Formulated Material IHE Testing

Detonation to Deflagration Transition (DDT) Testing

DDT testing is performed on IHE candidate materials to demonstrate that the HE will not transition to detonation in relevant to the material's application. The test requires high confinement to replicate a specific environment. The DDT test tube set up is shown in Figure 3. The scale of this experiment is extreme for a booster application; the conservatism is meant to bound main charge materials, which are used on a larger scale than that of booster materials.

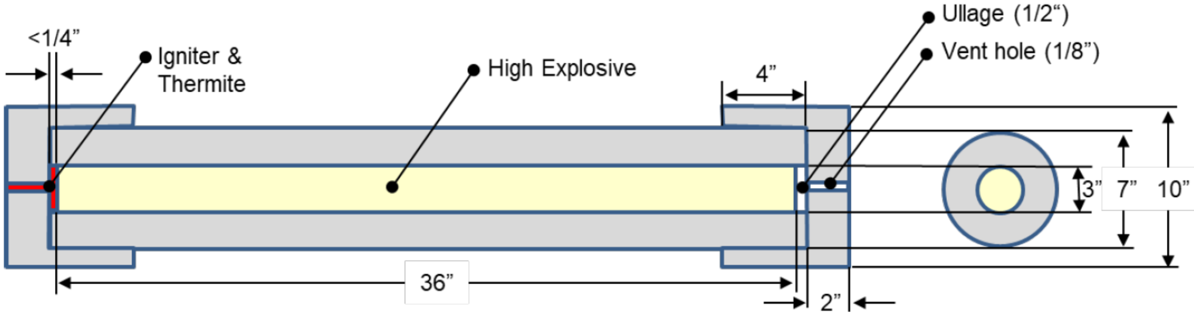

Figure 3. DDT test tube configuration [4]

Three full scale DDT tests were fielded on LLM-105 formulated material at LLNL's S300 Outside Firing Facility (OFF). One repetition of each test series (as defined by LLNL-TR-679331-REV-1) was completed:

- Slow cook-off: pressed to nominal density heated at a rate of 20°C/hr until self-ignition occurs (to determine material critical temperature, T_C)
- Thermite ignited nominal density brought to 10°C below the T_C and ignited using thermite glow plugs
- Thermite ignited hand-poured density (molding powder) brought to 10°C below the T_C and ignited using thermite glow plugs

The LLM-105 formulated material did not transition from deflagration to detonation on either of the production density tests. The DDT tube remained intact after completion of the production density slow cook off test (Figure 4) and the thermite ignited nominal density test (Figure 5). During the hand poured density test, the material underwent a detonation, which was determined by fragment analysis on the remnants of the DDT tube after the test (Figure 6). The fragments were compared to the fragments from an intentional detonation performed by Los Alamos National Laboratory (LANL) on TATB formulated material and found to be comparable. Pixel analysis of highspeed video was also performed to obtain an approximate horizontal wall velocity of 676 m/s.

Figure 4. DDT tube after LLM-105 formulation nominal density slow cookoff test

Figure 5. DDT tube after LLM-105 formulation nominal density thermite ignited test

Figure 6. Fragments of the LLM-105 formulated molding powder pour density DDT tube

Shock to Detonation Transition (SDT) Testing Summary

SDT testing is completed to define what shock stimulus will cause the HE to detonate. In the context of IHE testing, the purpose of SDT is to demonstrate that the IHE candidate material will not transition to detonation under the defined shock stimulus and defined shock duration at ambient temperature. The IHE test definition also requires that high temperature tests also be fielded, to demonstrate that the explosive is not overly sensitized, such as by a phase transition at elevated temperature. Note that to function in a weapon configuration, the HE must undergo SDT at some shock stimulus. The SDT test setup is shown in Figure 7. The conditions of the defined 1-D planar shock that a material must withstand without detonation to qualify as an IHE are:

At ambient temperature:

- 3.5 GPa for a duration of > 3.0 µs

- 5.3 GPa for a duration of > 0.5 µs

At high temperature:

Material is heated to 10°C below the T_C (determined experimentally through the slow cook-off DDT test) at a ramp rate of 20°C/hr

- 1.5 GPa for a duration of > 3.0 µs

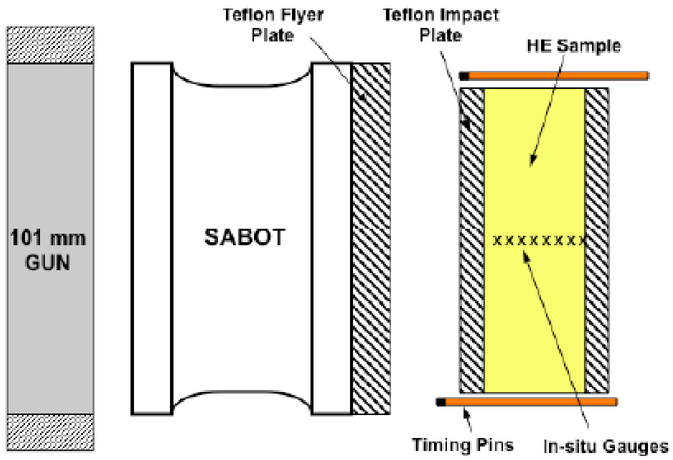

Figure 7. SDT test configuration [4]

Shock initiation and IHE qualification experiments on LLM-105 formulated material were performed to obtain in-situ pressure gauge data, run-distance-to-detonation thresholds, and IHE qualification results. The data from the SDT test series are displayed in Table 1.

Material	Temp [°C]	Density [g/cc]	Impact Velocity [m/s]	Pulse Duration [µs]	Impact Pressure [GPa]	Transition to Detonation [Y/N]
LLM-105 Formulation	Ambient	1.812	731	5.0	4.2	Yes**
LLM-105 Formulation	Ambient	1.812	896	0.5	5.7	Yes**
LLM-105 Formulation	Ambient	1.820	642	5.0	3.4	No
LLM-105 Formulation	Ambient	1.820	865	0.5	5.6	No
LLM-105 Formulation	Hot	1.820	416	>3	1.55	No
LLM-105 Formulation	Hot	1.820	423	>3	1.59	No

Table 1. SDT results of LLM-105 formulated material

* Velocity not measured so approximate velocity used based on powder load used

**Transitioned to detonation, experiment impact velocity resulted in a pressure exceeding that listed in the IHE Shock to Detonation (SDT) threshold

***Transitioned to detonation, experiment impact velocity resulted in a pressure exceeding that listed in the IHE Shock to Detonation (SDT) threshold and flyer separated

The three tests that transitioned to detonation fell outside the targeted velocity though provide useful data to establish a shock threshold. These three tests were over-tests, the HE is required to detonate upon some level of shock as noted earlier, and therefore do not disqualify this material as an IHE due to the SDT response.

Skid Testing

Skid Testing is primarily a worker safety test, intended to demonstrate that a billet of explosive material will not react with significant violence in a combined environment (drop and shear friction). The test is designed to simulate a more severe impact than what might occur during a handling accident. Using a pendulum system, a HE sample is struck against a gritted surface at varying angles. The LANL-designed test fixture is shown in Figure 8. A failure of the test is defined by the observation of “Reaction 2”, propagating/luminous flames visible on the high-speed video and cracks/visible damage on the HE part posttest. The IHE test definition defines three potential reactions, with reaction 0 and 1 yielding an acceptable test result. The criterion for each reaction is listed below:

- Reaction 0 is acceptable:
    - No visible smoke. No scorching of explosive surface. Video may show glowing abrasive particles from target surface.
- Reaction 1 is acceptable:
    - Visible smoke. Non-propagating, luminous ignition sites may be visible. Scorching of the explosive surface. No luminous flames are visible in highspeed videography from a side view.
- Reaction 2 is not acceptable:
    - Propagating, luminous flames are visible from high-speed videography. Postmortem examination reveals partial or complete disintegration of the explosive contact surface due to cracking induced by explosive reaction. [4]

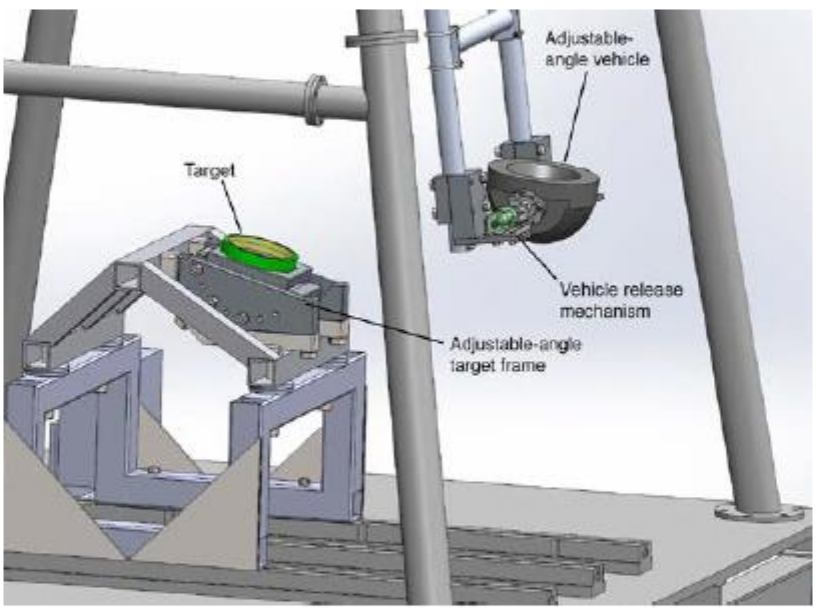

Figure 8. LANL-designed Skid test set up [4]

LLM-105 formulated material underwent Skid Testing at LANL. A total of 21 drop tests were conducted, including three repetitions at 12 feet and 45° angle, as is defined by the IHE test definition standard. The remaining 19 drops were performed at varying heights and angles. No light was observed from the high-speed camera at any of the drop heights/angles, which confirms that Reaction 2 did not occur during any of the drops. The data set from the complete Skid Test series completed is shown in Table 2.

Drop #	Impact Surface	Drop Height [ft]	Incident Angle [°]	Grit Size [μm]	Result
628	Clean Glass	6	45	N/A	No ignition
629	Gritty Glass	1	45	250-500	No ignition
630	Gritty Glass	1.5	45	250-500	No ignition
631	Gritty Glass	3	45	250-500	Visible smoke
632	Gritty Glass	6	45	75-150	Visible smoke
633	Gritty Glass	6	45	150-250	Visible smoke
634	Gritty Glass	6	45	250-500	Visible smoke
635	Gritty Glass	6	45	500-1000	Visible smoke
636	Gritty Glass	12	45	250-500	Visible smoke
637	Gritty Glass	12	45	250-500	Visible smoke
638	Gritty Glass	12	45	250-500	Visible smoke
639	Gritty Glass	6	14	250-500	Visible smoke
640	Gritty Glass	6	14	250-500	Visible smoke
641	Gritty Glass	6	14	250-500	Visible smoke
642	Gritty Glass	6	30	250-500	Visible smoke
643	Gritty Glass	6	30	250-500	Visible smoke
644	Gritty Glass	6	60	250-500	Visible smoke
645	Gritty Glass	6	60	250-500	Visible smoke
646	Gritty Steel	12	45	250-500	Visible smoke
647	Gritty Steel	12	45	250-500	Visible smoke
648	Gritty Steel	12	45	250-500	Visible smoke

Table 2. Skid Test details and results [5]

Bullet Impact Testing

Bullet Impact testing is performed on IHE material candidates to show that the HE will not react violently when impacted by a bullet (.50-caliber military ball round and .50-caliber armor piercing). This test is not intended to demonstrate that the HE will not react when impacted by any potential ammunition, but under the specific conditions set by the IHE test definition. To pass the Bullet Impact test, a HE may burn, even become completely consumed by burning, have smoke and visible light detected, and the assembly may become distorted. However, any further damage (i.e., a fragmented assembly) is considered a failure per the IHE test definition [4]. The Bullet Impact configuration is depicted in Figure 9. The IHE definition standard dictates that IHE materials must not react violently to bullet impact under these conditions:

- Ammunition: .50-caliber armor piercing or .50-caliber military ball round
- Muzzle velocity: standard for each type but must remain within 2,700 ft/sec to 3,050 ft/sec for a valid test [4]

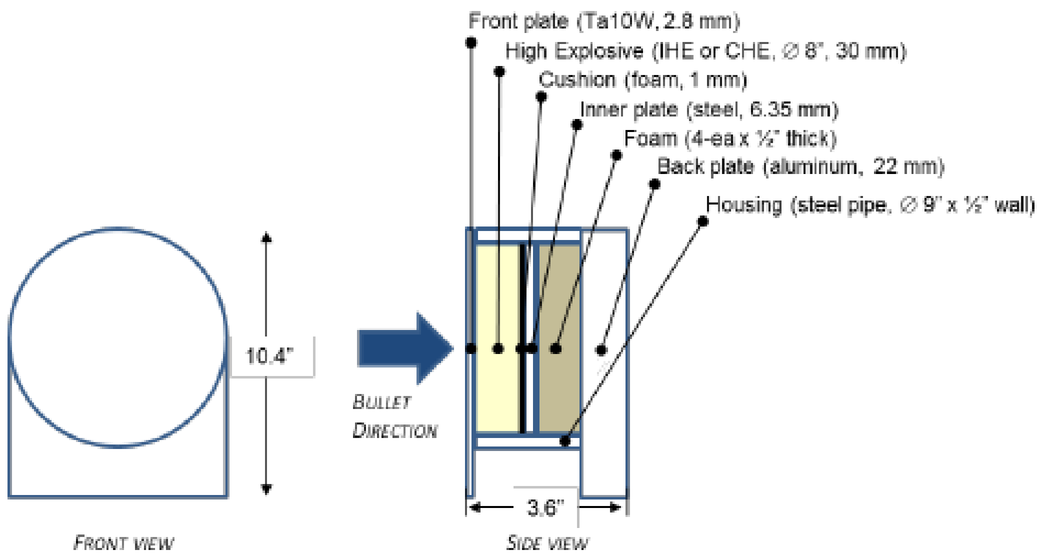

Figure 9. Bullet Impact test configuration [4]

Three repetitions of Bullet Impact Testing were performed on LLM-105 formulated material and the results are displayed in Table 3. The high-speed video of the test shows the bullet entering the assembly and material being ejected from the assembly, but the assembly is not fragmented and results in this material passing the Bullet Impact tests. The results from the experiment were comparable to the results from qualified IHE materials (TATB based formulation) performed previously in HEAF at LLNL in the same configuration [6] [7].

Projectile	Projectile Weight [g]	Average Velocity [ft/s]	Maximum Velocity [ft/s]	Pass/Fail
Armor Piercing	695.3	2835	2852	Pass
Ball Round	647.4	2778	2778	Pass
Ball Round	647.7	2934	2934	Pass

Table 3. Bullet Impact data

Conclusion

Overall, LLM-105 material formulated with elastomeric polymer has passed three of the four IHE material qualification tests. The material was put through a series of screening tests to determine if it would pass as an IHE material if tested against the entire IHE Qualification Standard prescribed in DOE-STD-1212-2019 Chapter 16. The material passed all IHE Qualification relevant tests in regard to Bullet Impact and SDT. A modified version of Skid Testing was performed by LANL where the material displayed IHE characteristics. During DDT testing, the LLM-105 material passed two trials of the production density pressed part DDT test. A detonation occurred during the pour density molding powder DDT test.

There are several follow on studies that the LLNL team would like to perform to investigate the detonation observed in the pour density DDT test. The first test to perform would be an intentional detonation study on LLM-105 formulated material with additional diagnostics to fully characterize the detonation behavior of LLM-105 formulated material in a DDT scenario.

In conclusion, for the booster applications currently being studied using LLM-105 formulated material, the HE appears to contain IHE safety characteristics for the intended use.

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