



## DBX-1 (copper(I)-5-nitrotetrazolate) reactions at sub-millimeter diameters

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### ABSTRACT

Continued motivation to reduce the amount of sensitive explosives used in devices has led to the need for a better understanding of the properties of energetic materials at reduced length scales. Additionally, the importance of minimizing the use of objectionable elements such as lead has driven research on suitable replacement materials. DBX-1 (copper(I)-5-nitrotetrazolate) has recently been developed as a promising replacement for lead azide. This research made use of high-speed streak camera photography to study the reaction properties of charges of DBX-1 at sub-millimeter diameters. The samples developed for study were cylindrical charges of DBX-1/dispersant contained in glass capillaries with sub-millimeter diameters. A laser (1064 nm) was used to ignite the charges. DBX-1 appears to undergo a Type I DDT (deflagration-to-detonation) process in which a plug of material develops, eventually shocking up to a detonation. The critical diameter for DBX-1 at  $1.6 \pm 0.1 \text{ g/cm}^3$  (58–66% dense), with glass confinement is less than 0.1 mm.

### 1. INTRODUCTION

For safety reasons, it is always desirable to reduce the amount of sensitive energetic materials (EMs) such as primary explosives in devices when possible. This has led to the need for a better understanding of the properties of energetic materials at reduced length scales. Primary explosives are the materials of choice for small-scale, low-energy use. While it has long been known that the growth-to-detonation lengths and the critical diameter for primary explosives must be quite small, there has been little work performed on measuring these properties.

There are two accepted mechanisms for the deflagration-to-detonation transition (DDT) in primary explosives [1-3]. Type I DDT is described as when the rising pressure of the combustion compresses unreacted material ahead of the combustion front and accelerates this compaction plug further into the

material until a shock builds and a detonation is achieved. Type II DDT is described as when the reaction progresses from conductive burning to convective burning, compressive burning, shock wave formation and finally a detonation transition.

In primary explosives, the DDT process occurs over such small length scales that it is only possible to study the process at small diameters. This work describes further application of a previously-developed [4, 5] experimental technique for preparing and studying sub-millimeter samples of primary explosives. The work described in this paper is on DBX-1 (copper(I)-5-nitrotetrazolate), which has recently been developed as a promising replacement for lead azide [6].

### 2. EXPERIMENTAL

The samples developed for study were cylindrical charges of DBX-1 (copper(I)-5-nitrotetrazolate) contained in micro-capillaries. The DBX-1 used in these experiments was used as obtained from Pacific Scientific Energetic Materials Co. (Chandler, AZ). The DBX-1 had been milled in isopropyl alcohol to reduce the average (volume-based) particle size to  $4.65 \mu\text{m}$  (90%  $< 9.32 \mu\text{m}$ , Microtrac).

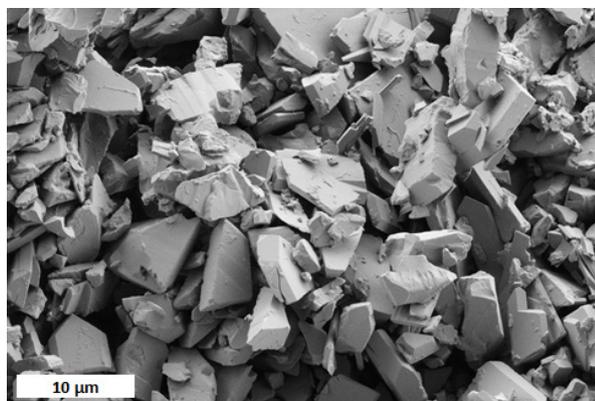
The process developed to prepare samples for these experiments involved dipping a microcapillary into a suspension consisting of the energetic material temporarily dispersed in a carrier fluid. The suspension would then flow into the microcapillary by capillary action. Capillaries were suspended vertically on microscope slides and allowed to dry overnight. The energetic material fell out of suspension and settled to the bottom of the microcapillary forming a cylindrical plug. Fluid surface tension and the capillary forces due to the small inner diameter prevented the suspension from leaking out of the end during drying.

Borosilicate glass micro-capillaries (Drummond Scientific Co., Broomall, PA) were used to confine the DBX-1. The capillaries had internal diameters of 0.099–0.480 mm. In order to fill the micro-capillaries,

it was necessary to formulate a suspension in which the DBX-1 would remain temporarily dispersed. If the particles were too well suspended in the carrier fluid they would adhere to the inner wall during drying rather than falling to the bottom to form a plug of material. Thus, the suspension stability time could not exceed the evaporation time of the carrier fluid. The final dispersion consisted of  $\sim 0.1$  g/mL of DBX-1 in mixed esters with 1.5–2.0% non-ionic dispersant. After all capillaries were filled and air-dried, they were dried in an oven for  $\sim 1$  hour at  $60^\circ\text{C}$ .

The density of the DBX-1/dispersant in the capillaries was calculated from careful mass measurements of the capillaries before and after preparation. Optical micrographs after filling were used for length measurements of the DBX-1 column. Only capillaries that contained greater than 0.5 mg of material were used to determine the average density of the DBX-1. The variance in densities below this mass was high due to measurement issues with low masses and short plug lengths. The density of DBX-1/dispersant in these capillaries was determined to be  $1.6 \pm 0.1$  g/cm<sup>3</sup> (58–66% dense).

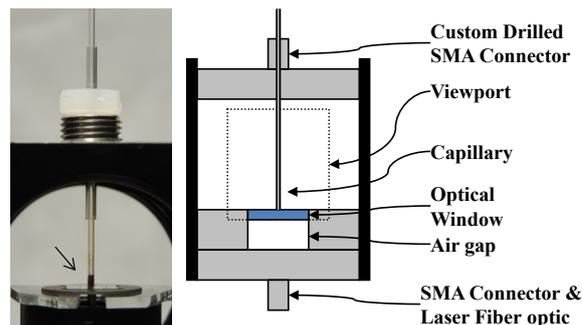
Scanning electron microscopy was used to examine the material removed from one of the microcapillaries after preparation (Figure 1). The DBX-1 particles are jagged, with both crystal facets and fractured surfaces visible. There is no obvious evidence of the dispersant, but there is no reason to believe that it was removed during sample preparation.



**Figure 1. Scanning electron micrograph of DBX-1/dispersant after removal from a microcapillary.**

The laser used for initiation was a two-output, fiber optic coupled, Nd:YAG laser with a wavelength of 1064 nm and a nominal pulse width of 12 ns. The laser was coupled to the sample with a 400- $\mu\text{m}$  core diameter, 0.39 NA fiber optic. An optical window was used to support the capillary and protect the face of the fiber optic cable. Due to the small sample sizes, a Nikon AF Micro Nikkor 105-mm 1:2.8 macro zoom lens was placed between the sample and the slit of the 16-bit dynamic range streak camera to achieve the necessary magnification. The small size of the samples resulted in a very low amount of light produced during reaction: therefore all samples were

fired in minimal ambient light. The experiment used to fire the samples is shown in Figure 2.



**Figure 2. (left) Photograph of a microcapillary containing a short column of DBX-1 ready to be fired. (right) Diagram of the elements of the firing experiment.**

The two outputs of the laser allowed for correlation of the energy at the output of the second fiber optic with that of the fiber optic used for initiation. The fluence (energy/area) was estimated by measuring the spot size of the laser at an output energy of 9 mJ to be  $\sim 0.8$  mm. The fluence on each sample required for initiation ranged from 0.5 to 15 J/cm<sup>2</sup>, with higher fluences required for the smaller samples, presumably due to more pronounced thermal losses at smaller diameters.

The streak images recorded in these experiments were analyzed with edge-finding software and a data analysis method described elsewhere [7]. The reactions appear in the streak images as a bright streak against a dark background. The software extracts the line of a reaction from a streak image and exports it as a series of position versus time data points. Static images were taken with the streak camera to determine the position of the top and bottom positions of DBX-1 in each microcapillary. A static image of a 100-line-per-inch Ronchi rule was taken at the same magnification of the samples for length scale calibration for the software. The time information for each streak image was recorded internally by the streak camera and read from the image file header by the edge-finding software. For steady-state reactions, a linear least square regression was used to calculate the reaction velocity. Error in the velocity is reported as plus-or-minus the standard error of the fit plus 5%.

### 3. RESULTS AND DISCUSSION

A total of 60 experiments were conducted with the goal of recording steady detonation in multiple capillaries at each of the six diameters used. Detonation was recorded in 18 of these 60 experiments. Of the experiments where detonation was not recorded were three classes of results: the timing was such that no data were recorded, the capillary did not ignite, or the reaction did not grow to detonation. Failure of growth to detonation was more prevalent in shorter columns, suggesting a self-confinement effect. In many of the experiments with short column lengths, the luminous reaction pushed deflagrating material up the length of the

microcapillary, but growth to detonation did not occur. The results from the 18 experiments that detonated are shown in Table 1 and Figure 3.

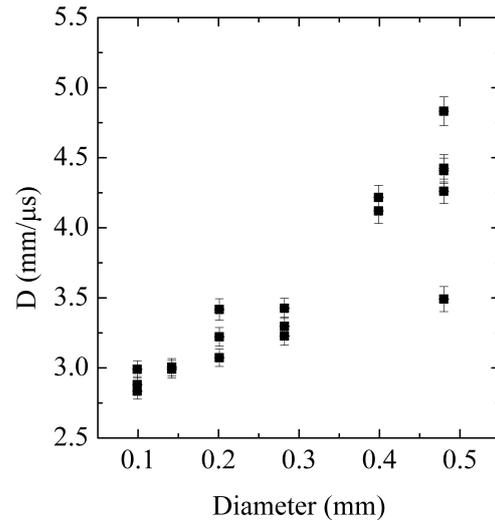
There is an obvious diameter effect in the data in which the detonation velocity decreases with diameter. The typical empirical form for data analysis of diameter effect data is:

$$D(R) = D_{\infty} \cdot \left( 1 - \frac{A}{R - R_C} \right), \quad [1]$$

where  $D(\infty)$  is the detonation velocity of an infinite radius charge at the same density,  $R$  is the charge radius in question,  $A$  is a length parameter,  $R_C$  represents the value of the asymptote corresponding to the failure radius, and  $D(R)$  is the detonation velocity of a charge of radius  $R$ . [6] This equation represents a decrease in detonation velocity with decreasing charge radius, and asymptotically approaching zero at the failure radius. The data for DBX-1 do not fit this form and we believe that additional data at both larger and smaller diameters would have to be collected to attempt a fit. Despite our inability to perform a fit and based on these results, the critical diameter for DBX-1 at  $1.6 \pm 0.1 \text{ g/cm}^3$  (58–66% dense), with glass confinement is less than 0.1 mm.

**Table 1. DBX-1 detonation velocity results for all microcapillaries that detonated.**

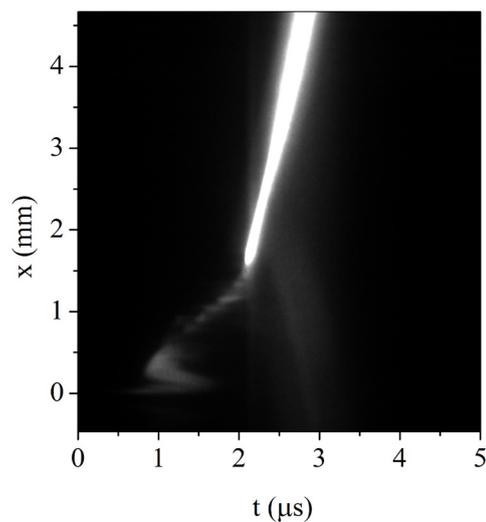
Capillary Diameter	Detonation Velocity	Column mass	Column height
mm	mm/ $\mu$ s	mg	mm
0.480	$3.492 \pm 0.090$	1.31	4.50
0.480	$4.832 \pm 0.102$	1.42	3.99
0.480	$4.425 \pm 0.098$	1.12	3.50
0.480	$4.407 \pm 0.090$	2.59	8.80
0.480	$4.261 \pm 0.087$	3.65	11.84
0.399	$4.120 \pm 0.088$	1.55	7.51
0.399	$4.216 \pm 0.087$	1.41	7.04
0.282	$3.425 \pm 0.072$	0.96	9.61
0.282	$3.277 \pm 0.068$	0.94	9.36
0.282	$3.228 \pm 0.066$	0.92	9.43
0.201	$3.222 \pm 0.066$	0.40	7.65
0.201	$3.073 \pm 0.062$	0.82	16.44
0.201	$3.417 \pm 0.077$	0.18	2.90
0.142	$3.005 \pm 0.062$	0.25	9.12
0.142	$2.991 \pm 0.062$	0.17	5.43
0.099	$2.881 \pm 0.059$	0.34	23.5
0.099	$2.990 \pm 0.061$	0.14	7.80
0.099	$2.834 \pm 0.0574$	0.24	17.02



**Figure 3. Graph of detonation velocity as a function of DBX-1 diameter in glass confinement.**

A variety of different qualitative reaction results were obtained during this experiment series. Due to the low light levels, many of the streak images had very low contrast. The streak images included in this paper have all been processed for the reader, but only unmodified streak images were used in the data extraction process.

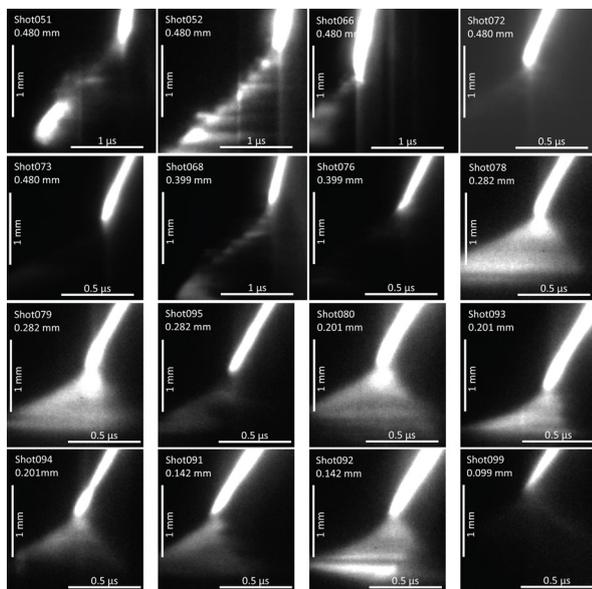
Growth to detonation is seen in the streak image in Figure 4. The interface between the bottom of the capillary and the laser window is at 0 mm and the transition occurs at  $\sim 1.5$  mm after a luminous, but dim deflagration. This is an example of Type I DDT in which an accelerative reaction front compresses a plug of material that eventually shocks up to a detonation.



**Figure 4. Streak image of ignition and growth to detonation in DBX-1. Capillary diameter: 0.399 mm, D:  $4.120 \pm 0.088 \text{ mm}/\mu\text{s}$ , DBX-1 mass: 1.55 mg, column height: 7.51 mm.**

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Every experiment where the DDT process was captured is shown in Figure 5. Additional evidence for Type I DDT can be seen in these images. Characteristic of this process is that at the base of the bright linear detonation, there is evidence of plug formation, seen as a narrow discontinuity. It is the leading edge of this plug that coincides with the initial formation of the shock front that causes the detonation transition. This is especially pronounced in the image for Shot095 in the second from bottom row.



**Figure 5.** Every image that was captured of the DDT process.

A cursory comparison of the DDT process in DBX-1 with lead azide under similar preparation conditions [5] suggests that the DDT length in DBX-1 is 2–3 times larger than that of lead azide. This could result from physical differences in the powders such as density and particle size. A more likely explanation is that the decomposition kinetics differ in a simple molecule such as lead azide than the slightly more complicated DBX-1.

#### 4. CONCLUSIONS

Sub-millimeter cylindrical charges of DBX-1 were prepared using a dispersion of the material in a non-solvent. This process is a convenient technique for preparation of small-diameter samples, provided that high-density material is not required. This technique is of particular interest for primary explosives due to the short length scales of DDT in these sensitive explosives.

DBX-1 prepared in this manner appears to undergo a Type I DDT process in which a plug of material develops, eventually shocking up to a detonation. A diameter effect was observed, but a fit to the typical empirical form was not possible with this incomplete data set. The critical diameter for DBX-1 at  $1.6 \pm 0.1 \text{ g/cm}^3$  (58–66% dense), with glass confinement is less than 0.1 mm.

#### 5. REFERENCES

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