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CRITICAL DETONATION THICKNESS IN VAPOR-DEPOSITED PENTAERYTHRITOL TETRANITRATE (PETN) FILMS

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Abstract. The use of physical vapor deposition is an attractive technique to produce microenergetic samples to study sub-millimeter explosive behavior. Films of the high explosive PETN (pentaerythritol tetranitrate) were deposited through vacuum thermal sublimation. Deposition conditions were varied to understand the effect of substrate cooling capacity and substrate temperature during deposition. PETN films were characterized with surface profilometry and scanning electron microscopy. Detonation velocity versus PETN film thickness was analyzed using a variation of the standard form for analysis of the diameter effect. Results were compared with previous work conducted on PETN films deposited with lower substrate cooling capacity. Seemingly subtle variations in PETN deposition conditions led to differences in detonation behaviors such as critical thickness for detonation, detonation velocity at “infinite” thickness, and the shape of the critical thickness curves.

Keywords: Microenergetics, microdetonics, critical diameter, critical thickness.

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INTRODUCTION

Critical detonation thickness within explosives is analogous to the more commonly described critical diameter [1, 2]. Despite its widespread use, there has been no literature on critical detonation phenomena in high-density pure PETN (pentaerythritol tetranitrate) until recently when vapor deposition of PETN allowed sufficiently small samples to be made [3]. This paper presents an extension of that work, evaluating the effects of different deposition conditions on critical thickness curves.

The general approach of our work on microenergetics is to manufacture test articles that have been fabricated such that a critical length dimension is sub-millimeter. Samples with these small length scales allow optical diagnostic access to a large fraction of the explosive and present an alternative experimental approach to traditional pressing of pellets for investigations into small-scale explosive phenomena.

Microstructural aspects of explosives such as particle size and porosity distribution are known to affect detonation [4]. During processes such as physical vapor deposition, changes in deposition parameters such as deposition rate and substrate temperature can affect microstructure in deposited films. Thus it follows that by varying deposition conditions, microstructure in explosives can be varied, and a better understanding of microstructural effects on detonation can be established. In this paper we present work showing that seemingly subtle changes in deposition conditions can result in significant variations in detonation behavior in vapor-deposited PETN.
**EXPERIMENTAL**

Films of PETN were deposited by vacuum thermal sublimation [5] in a custom deposition system on 10×30×0.5 mm fused silica substrates. A shadow mask was used to define two lines of PETN, each 1.00 mm wide, on each substrate. Different film thicknesses were achieved by changing the number of depositions and by adjusting the amount of PETN added to the effusion cell for the final depositions. A cooled substrate holder was used during the deposition process. Increases in coolant flow and area of cooling have been made since previous work [3], which motivated comparisons in resultant film properties. A variety of substrate conditions were examined related to three variables: cooling capacity of the substrate holder, substrate contact with the substrate holder, and temperature of the substrate holder.

PETN film thickness was determined from surface profiler (Dektak 8, Veeco) measurements at 13 locations along each PETN line. The average thickness of the center 100 μm of each measurement was used to calculate the average thickness along the length of the entire line. A film from each deposition condition was snapped to expose the cross-section (Fig. 1). Scanning electron microscopy (Supra 35VP SEM, Zeiss) was conducted to evaluate qualitative differences in film microstructure.

PETN film detonation velocity was determined using Sandia’s critical thickness experiment [3]. The PETN lines on each substrate were confined by a fused silica lid with 100 μm core optical fibers positioned in seven laser-machined holes along the length of each PETN line. Epoxy (Barco Bond MB-185, Astro Chemical Co., Inc.) was used to affix the lid and exclude air from the assembly. A line wave generator was used to ignite the lines. Previous experiments have shown no shock interactions between the two PETN lines and no detectable ignition transient. The detonation light transmitted from the optical fibers on each PETN line was detected with a silicon photodetector (DET210, Thorlabs) and recorded using a digital oscilloscope. Linear fits to the resultant position-time data were used to determine the detonation velocity.

**RESULTS AND DISCUSSION**

Detonation velocity versus inverse half thickness curves are constructed from the data and analyzed using the form:

\[
D(t_h) = D(\infty) \left[ 1 - \frac{1}{t_h} \left( \frac{A}{1 - \frac{t_h}{t_{hc}}} \right) \right],
\]

where \(D(t_h)\) is detonation velocity at a given explosive film half thickness (mm/μs), \(D(\infty)\) is the detonation velocity at infinite thickness (mm/μs), \(A\) is a length parameter specific to the explosive (mm), \(t_h\) is the explosive half thickness (mm), and \(t_{hc}\) is a length parameter equated to “critical half thickness” (mm). This form is derived from standard diameter effect analysis [1], with explosive radius replaced with explosive half thickness. In data sets that include detonation failure, the highest thickness point that failed is included in the analysis, using 0 mm/μs as the velocity. A summary of the deposition conditions and critical thickness curve values is shown in Table 1.
Table 1. Summary of deposition conditions and effects on critical thickness curve parameters.

<table>
<thead>
<tr>
<th>Substrate Cooling Capacity</th>
<th>Substrate Contact</th>
<th>Substrate Holder Temp. °C</th>
<th>Detonation Velocity at Infinite Thickness, D(∞) mm/μs</th>
<th>Critical Thickness, t_c mm</th>
<th>Length Parameter, A mm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Poor</td>
<td>N/A</td>
<td>7.82 ± 0.05</td>
<td>0.131 ± 0.003</td>
<td>0.0002 ± 0.0001</td>
<td></td>
</tr>
<tr>
<td>Good</td>
<td>Poor</td>
<td>7.72 ± 0.10</td>
<td>0.107 ± 0.014</td>
<td>0.0006 ± 0.0005</td>
<td></td>
</tr>
<tr>
<td>Good</td>
<td>Good</td>
<td>7.64 ± 0.28</td>
<td>0.133 ± 0.000</td>
<td>0.00004 ± 0.00004</td>
<td></td>
</tr>
<tr>
<td>Good</td>
<td>Good</td>
<td>7.66 ± 0.28</td>
<td>0.142 ± 0.000</td>
<td>0.0001 ± 0.00004</td>
<td></td>
</tr>
</tbody>
</table>

Figure 2. The effect of substrate holder cooling capacity on the critical thickness curve. PETN deposited with good substrate cooling capacity exhibits a lower detonation velocity at infinite thickness (lower density) and has a smaller critical thickness.

The effect of substrate holder cooling capacity is shown in Fig. 2. This comparison is made between the present work and previous work with PETN in 0.80 mm lines (as opposed to 1.00 mm), deposited with good substrate contact, but with less effective substrate holder cooling capacity [3]. The higher substrate holder cooling capacity resulted in material with a lower detonation velocity at infinite thickness, a lower critical thickness, and a larger length parameter, A (more heterogeneous character).

Substrate holder cooling capacity plays a role in how effectively heat is removed from the growing PETN film during deposition. Heating of the film results from both radiative transfer from the effusion cell as well as from heat of condensation liberated as the PETN vapor condenses on the substrate. It is likely that the higher detonation velocity at infinite thickness in the data labeled “poor substrate cooling capacity” resulted from higher density that may have resulted from higher thermal mobility in PETN admolecules. Deposition rate is likely to also have been important, but was not measured. With increasing deposition rate, there is less time available for an admolecule to migrate to lower energy (higher density) positions in the crystal lattice before being immobilized by additional deposited PETN. Thus, the smaller critical thickness and larger length parameter in the data with good substrate cooling capacity may represent a more heterogeneous explosive with lower density in which detonation failure is gradual due to distributed larger scale porosity.

The effect of substrate temperature is shown in Fig. 3. This comparison is made between two sets of PETN films deposited with good substrate cooling capacity and good substrate contact, but with different substrate temperatures of 10°C and 22°C. There is no significant difference in detonation velocity at infinite thickness or in length parameter, but the material deposited at 10°C has a smaller critical thickness. As with the argument above, this may be the result of lower thermal mobility of admolecules and resulting porosity that supports detonation in thinner films.
Figure 3. The effect of substrate holder temperature on the critical thickness curve. PETN deposited with a lower substrate holder temperature exhibits a smaller critical thickness, but has a similar detonation velocity at infinite thickness.

Scanning electron microscopy reveals no obvious differences in PETN microstructure among the films deposited at different conditions. Cross-sections of the films show polycrystalline columnar growth with similar scales of the individual needle-like crystals with sub-micron widths and lengths of several tens of microns. There appears to be a higher level of porosity in the PETN prepared with good substrate holder cooling capacity and at 10°C, but further work is necessary to quantify the porosity content in the different PETN films. The differences in detonation behavior in these deposited films is likely due to subtle variations in density, microstructure, and porosity distribution that arise from variations in deposition conditions.

CONCLUSIONS

Subtle changes in PETN deposition conditions resulted in dramatic effects in critical thickness curves. This includes lower detonation velocity at infinite thickness with higher substrate cooling capacity, and higher critical thickness with higher substrate temperature. While it is logical to conclude that these changes resulted from changes in microstructure (grain size, porosity distribution, density, etc.) that were affected by deposition conditions, no substantial differences were evident in micrographs. This suggests that future work should use sensitive methods to analyze microstructure to construct preparation-structure-property relationships in deposited PETN films.

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