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Sandia National Laboratories

Critical detonation thickness in vapor-deposited hexanitroazobenzene (HNAB) films with different preparation conditions

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11th Workshop on Pyrotechnic Combustion Mechanisms,
Colorado Springs, CO,
July 12, 2014.
15th International Detonation Symposium,
San Francisco, CA,
July 13–18, 2014.

U.S. DEPARTMENT OF ENERGY

NNSA National Nuclear Security Administration

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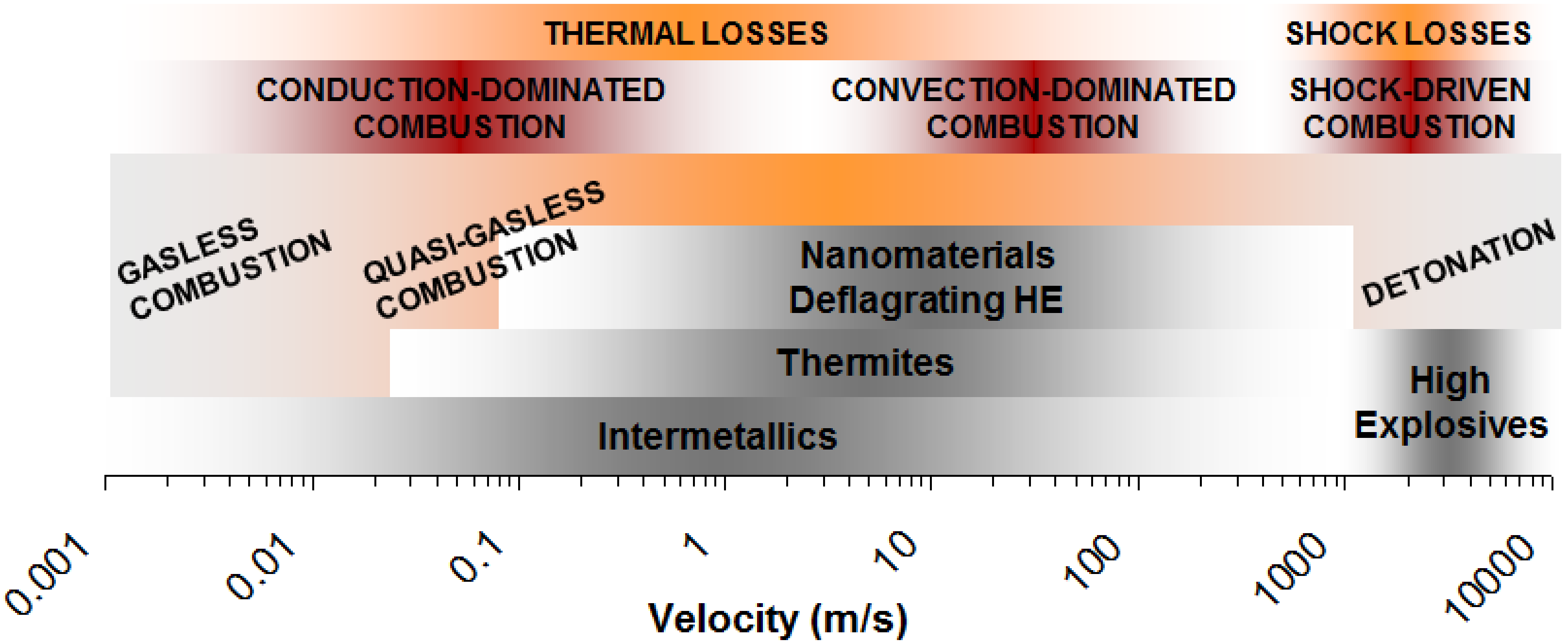
Energetic Material Considerations

- Energetic material reaction velocities span six orders of magnitude
- Material characteristics related to loss mechanisms (thermal or shock)
- Intrinsic material properties define failure
  - Energy density, velocity, critical diameter…



Tappan, A.S., "Microenergetics: Combustion and Detonation at Sub-Millimeter Scales," *15th APS Topical Conference on the Shock Compression of Condensed Matter*, Kohala Coast, HI, June 24-29, 2007.

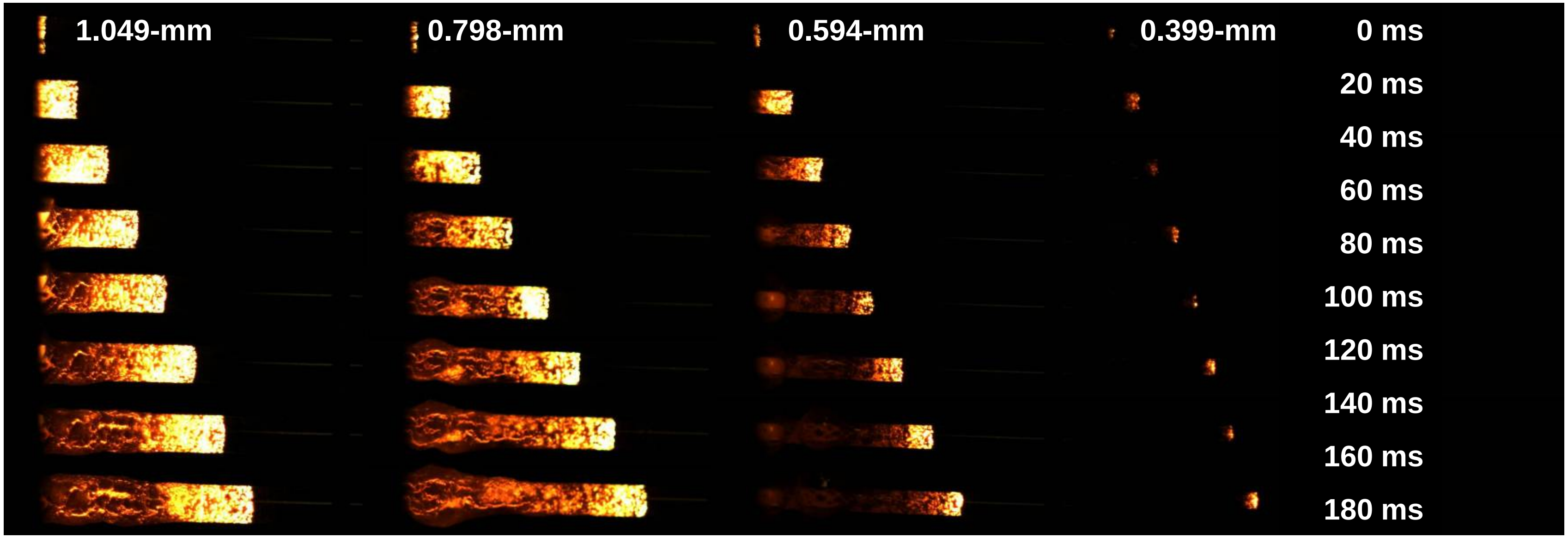
Reaction Front Comparison 75:25 Ti/2B-Ni/Al

- Decreasing diameter caused smaller reaction zone, slight velocity decrease

Tappan, A.S., Groven, L.J., Miller, J.C., and Puszynski, J.A., "Combustion Synthesis in Gasless Pyrotechnics at Millimeter Geometries," *Europyro 2007 and 34th International Pyrotechnics Seminar*, Beaune, France, October 8-11, 2007, pp. 69-79.

Reaction Front Comparison

- Mixture 4 (75:25 Ti/2B-Ni/Al)
- Decreasing capillary diameter caused smaller reaction zone, slight velocity decrease



Tiled images from mixture 4 (75:25 Ti/2B-Ni/Al) showing combustion in the four diameters.

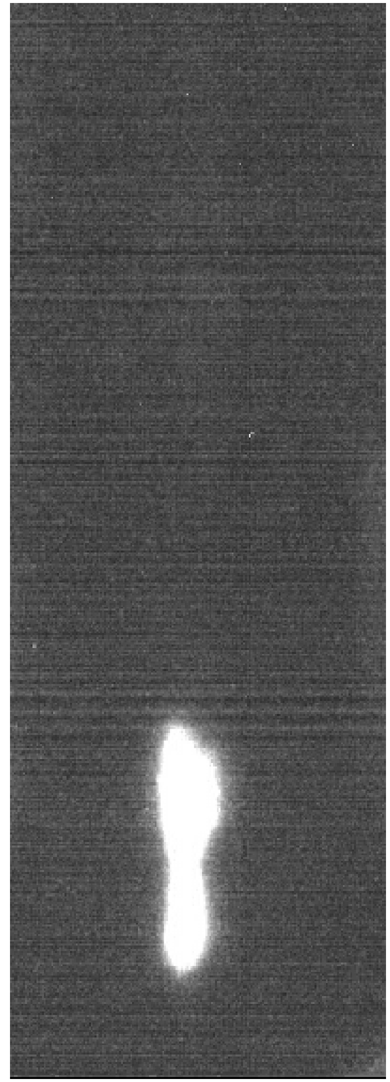
Nanocomposite Thermite Microchannel with Piston

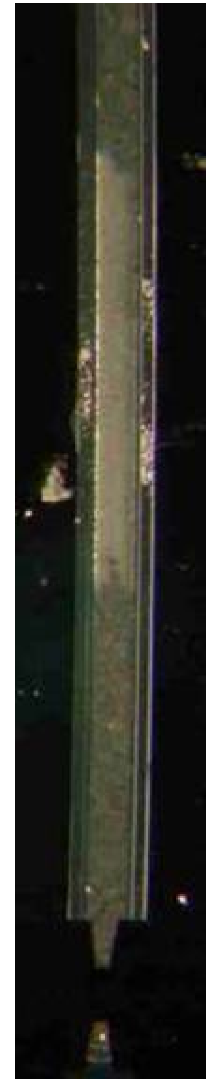
- 300 µm wide × 100-µm deep rectangular channel
- Device was powder-filled with nanocomposite thermite, 40:60 Al:MoO3, Steve Son
- Piston was 300 × 100 µm × 2 mm and weighed 313 µg
- 500,000 fps (1/2 µs)
- 2 µs exposure time

Tappan, A.S., "Microenergetics: Combustion and Detonation at Sub-Millimeter Scales," *15th APS Topical Conference on the Shock Compression of Condensed Matter*, Kohala Coast, HI, June 24-29, 2007.

Nanocomposite Thermite Microchannel without Piston

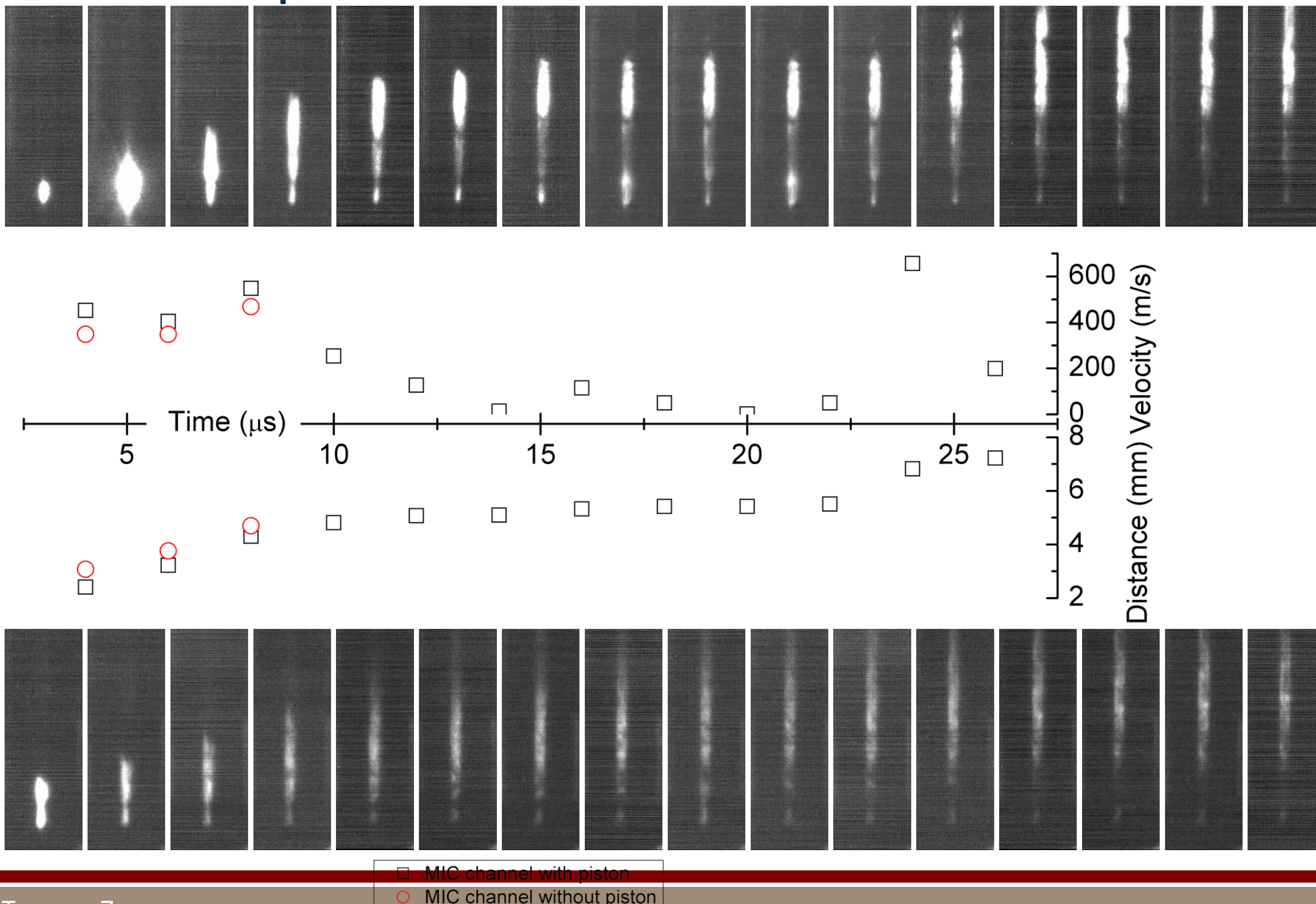
- 300 µm wide × 100-µm deep rectangular channel
- Device was powder-filled with nanocomposite thermite, 40:60 Al:MoO_3, Steve Son
- No piston and visible air gap
- 500,000 fps (1/2 µs)
- 2 µs exposure time





Tappan, A.S., "Microenergetics: Combustion and Detonation at Sub-Millimeter Scales," *15th APS Topical Conference on the Shock Compression of Condensed Matter*, Kohala Coast, HI, June 24-29, 2007.

Comparison of Nanocomposite Thermite Microchannels



Introduction to Detonation Diameter Effects

- Detonation failure
  - Occurs when size (diameter or thickness) of explosive is decreased
  - When surface losses dominate behavior
- Data for small-scale behavior of high-density pure explosives are scarce
  - Difficult to prepare small-scale samples
  - Failure length scales are often sub-millimeter

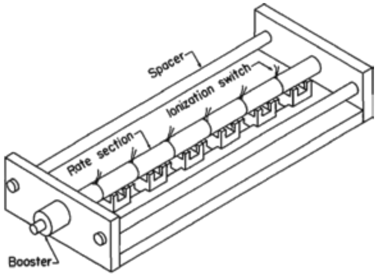
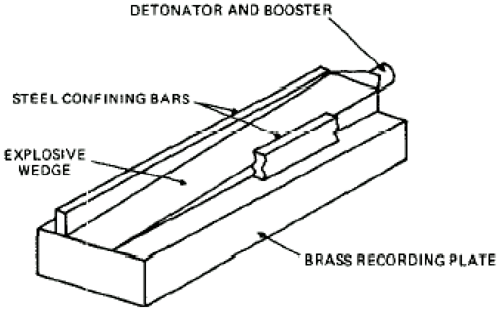
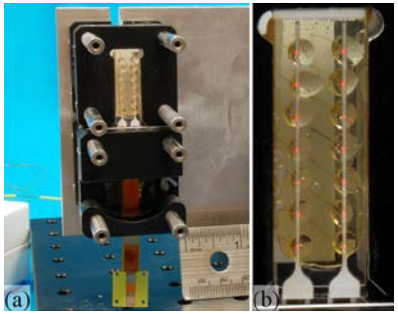


Fig. 1. Schematic of a typical rate-stick assembly.

Rate stick experiment.



Detonation failure experiment.



Critical thickness experiment.

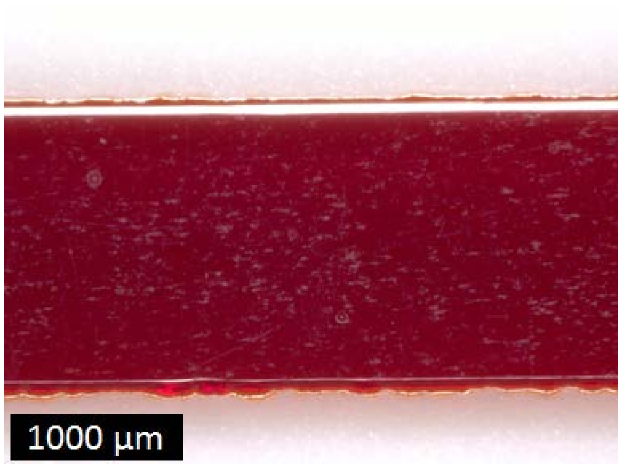
Campbell, A.W. and Engelke, R., "The Diameter Effect in High-Density Heterogeneous Explosives," 6th Symposium (International) on Detonation, Coronado, CA, August 24-27, 1976, pp. 642-652.

Gibbs, T.R. and Popolato, A., LASL Explosive Property Data, Detonation Failure Thickness, pp. 289-290. Berkeley, Los Angeles, London: University of California Press, 1980.

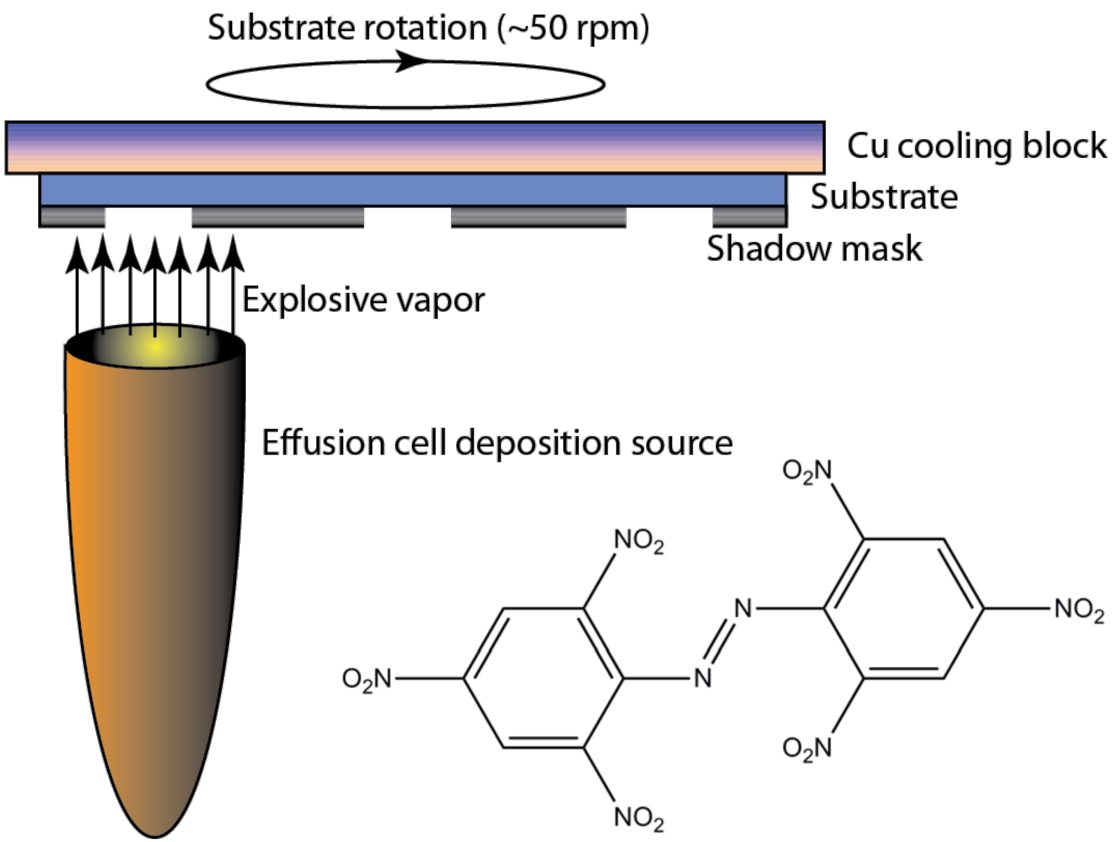
Tappan, A.S., Knepper, R., Wixom, R.R., Marquez, M.P., Miller, J.C., and Ball, J.P., "Critical Thickness Measurements in Vapor-Deposited Pentaerythritol Tetranitrate (PETN) Films," 14th International Detonation Symposium, Coeur d'Alene, ID, April 11-16, 2010.

HNAB physical vapor deposition

- Polycarbonate substrates
- Amorphous HNAB films
- 100% dense

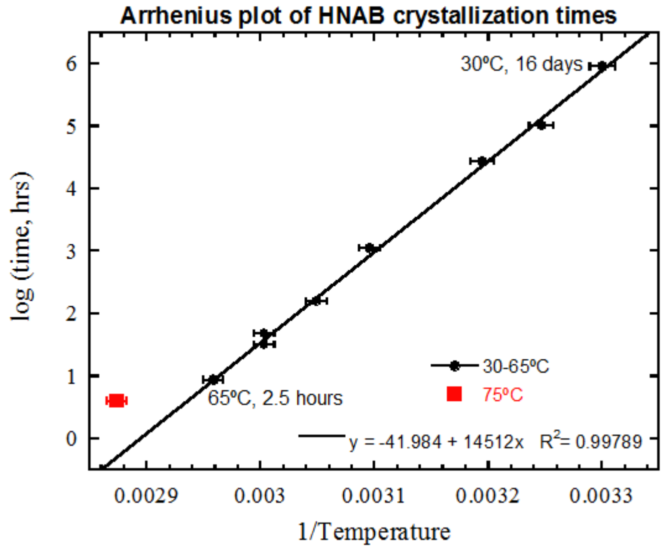


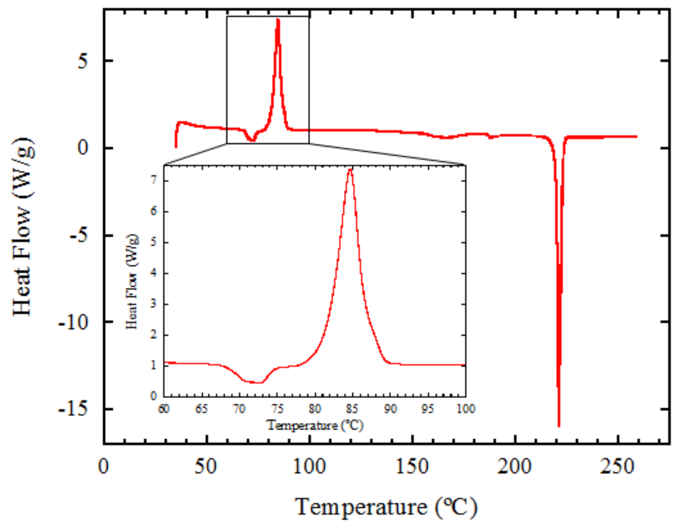
Optical micrograph of as-deposited HNAB.



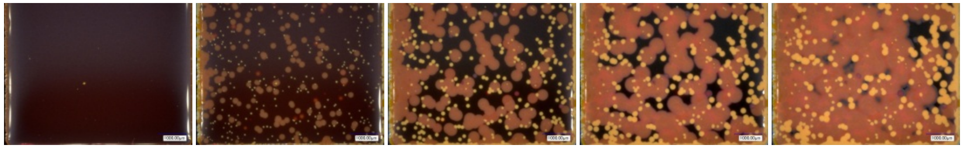
HNAB crystallization

- Amorphous HNAB films crystallize over time
- Pronounced difference in crystallization above glass transition temperature (T_g, ~70 °C)





DSC data from an amorphous HNAB film heated from 40–250 °C at 5 °C/min.

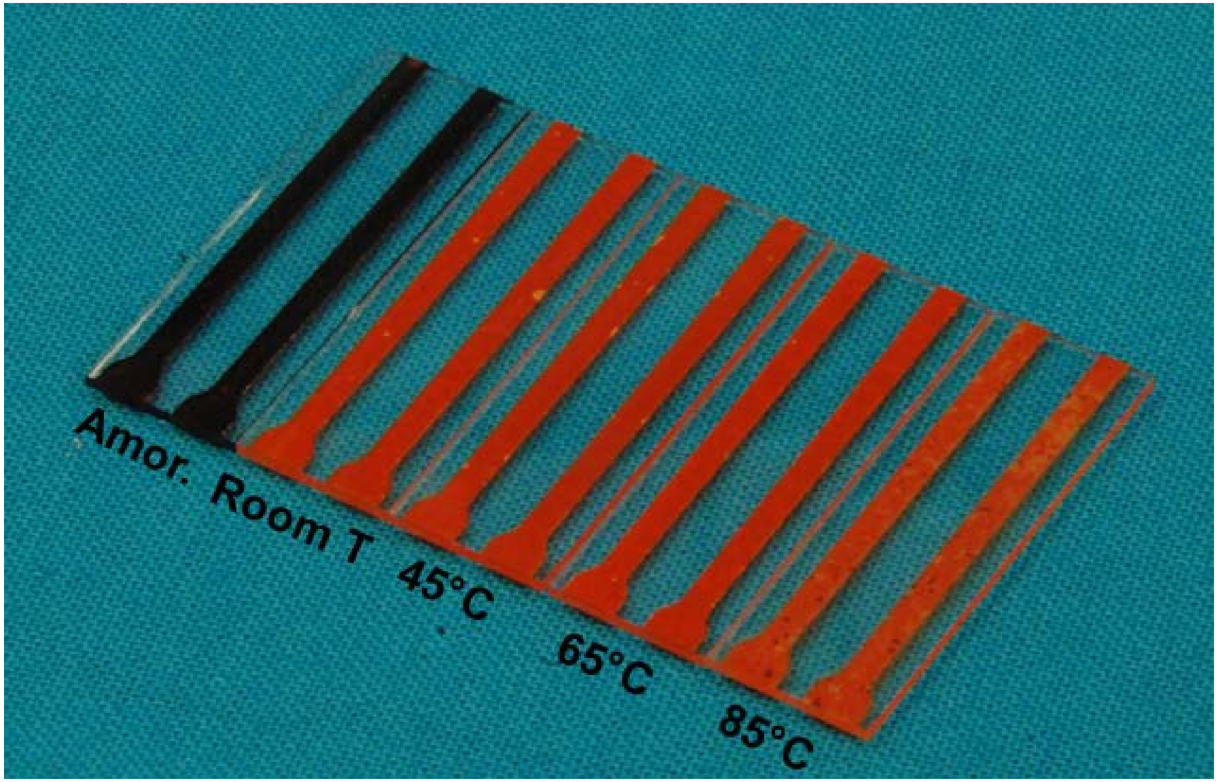


Time-lapse of HNAB crystallization, 65 °C, 24 min./image.

Knepper, R., Browning, K., Wixom, R.R., Tappan, A.S., Rodriguez, M.A., and Alam, M.K., "Microstructure Evolution during Crystallization of Vapor-Deposited Hexanitroazobenzene Films," *Propellants, Explosives, Pyrotechnics*, vol. 37, pp. 459 – 467, 2012.

HNAB samples

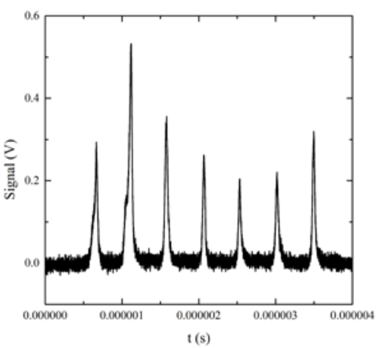
- Polycarbonate substrates
  - 1 × 3 cm
- Four crystallization conditions
  - Amorphous
  - Room temperature
  - 65 °C
  - 85 °C

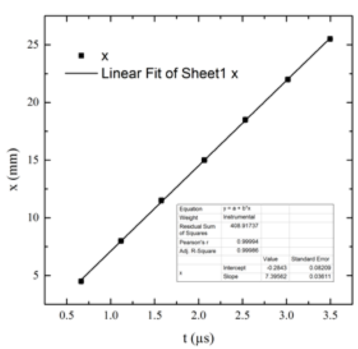


Photograph of five HNAB films on polycarbonate. Amorphous film on left and increasing crystallization temperature to right.

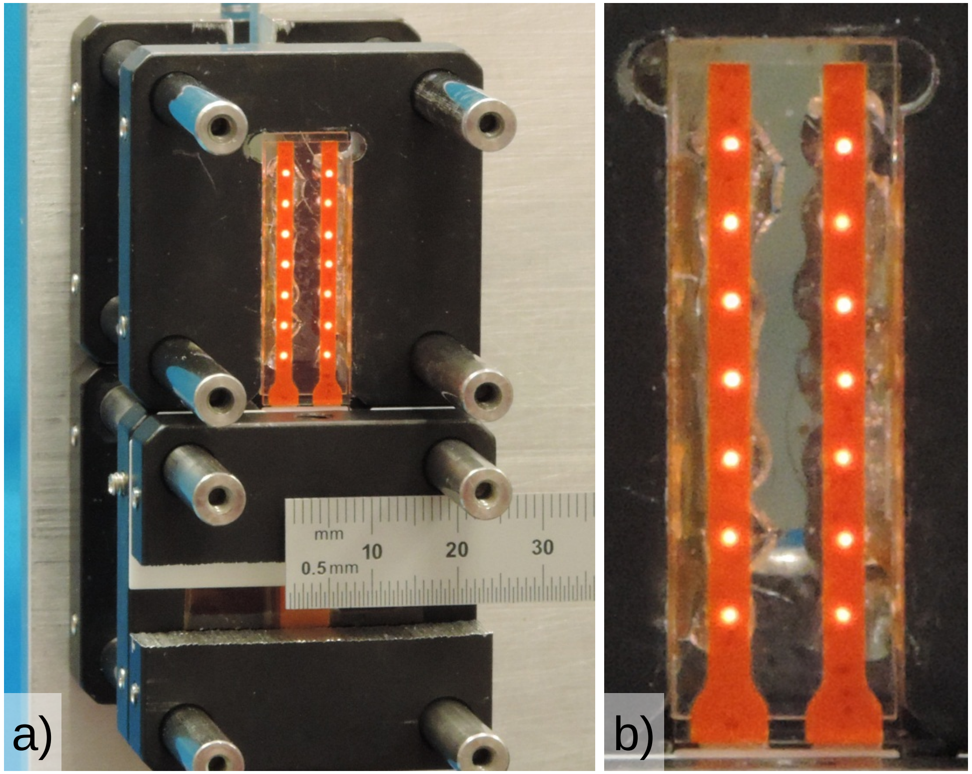
Critical detonation thickness experiment

- Two experiments (HNAB lines) each shot
- Optical fibers deliver detonation light to photodetector



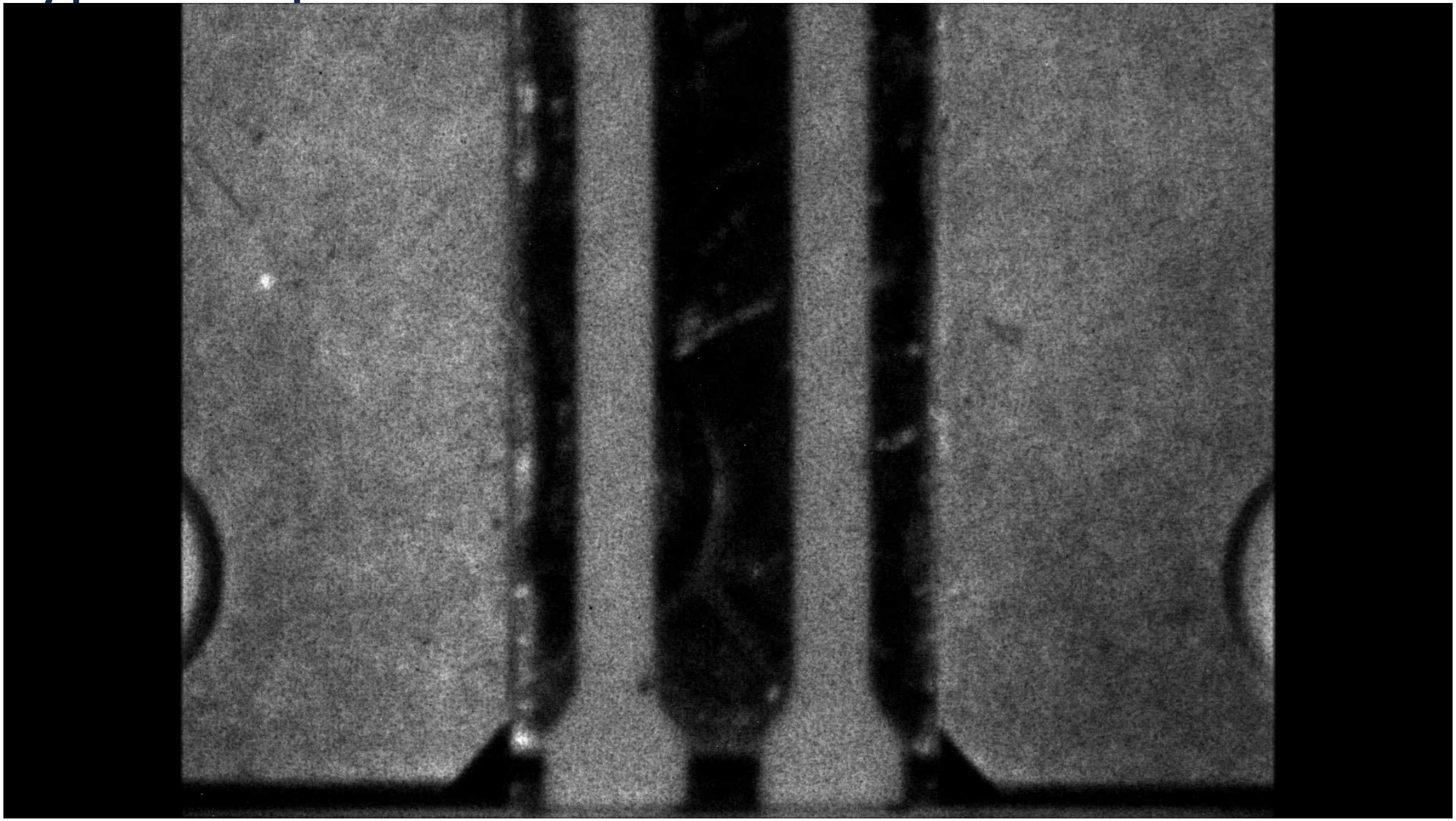


Optical fiber data is used to produce a linear fit to position versus time, where the slope is the velocity.



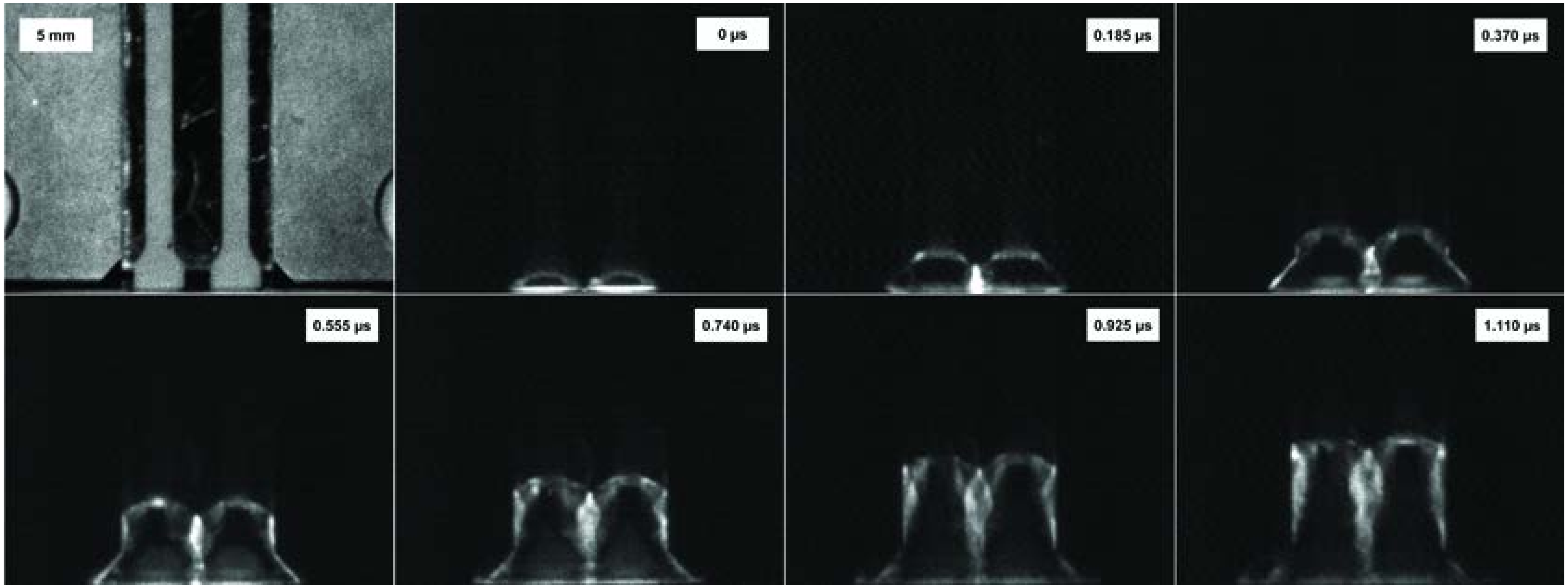
Photographs of critical detonation thickness experiment. Optical fibers illuminated to highlight position.

Framing camera images of a typical experiment



Framing camera images of detonation in the two lines of a single HNAB sample crystallized at 45 °C. These images were taken at 5.4 million frames-per-second (1/185 ns) with an exposure time of 15 ns.

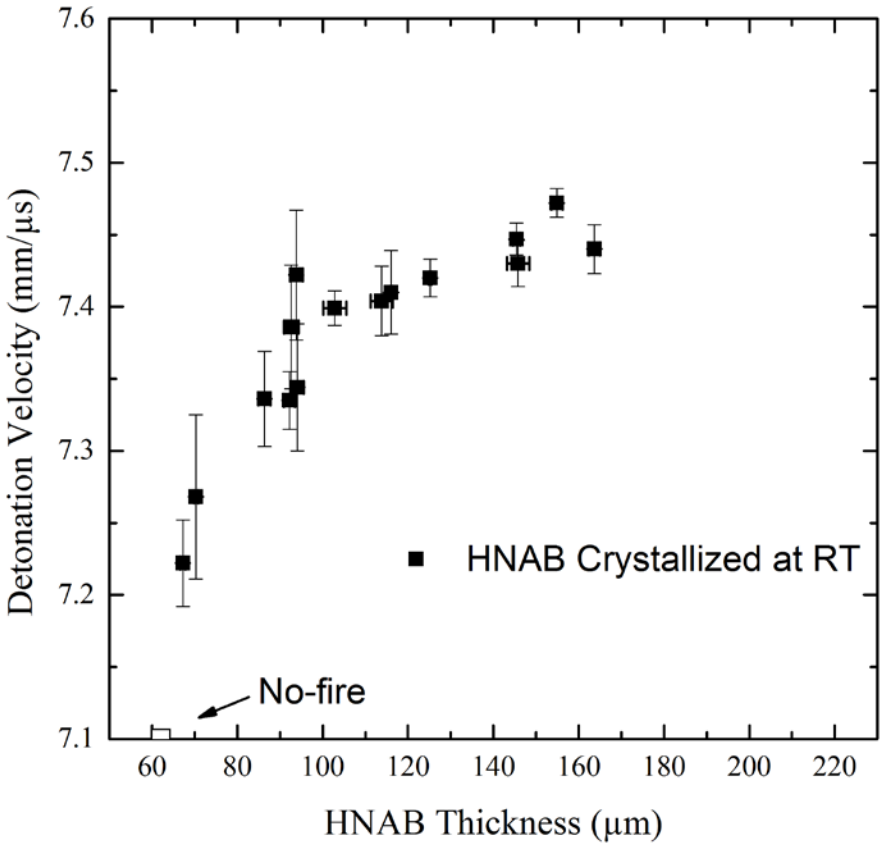
Framing camera images of a typical experiment



Framing camera images of detonation in the two lines of a single HNAB sample crystallized at 45 °C. These images were taken at 5.4 million frames-per-second (1/185 ns) with an exposure time of 15 ns.

Failure thickness for HNAB crystallized at room temperature

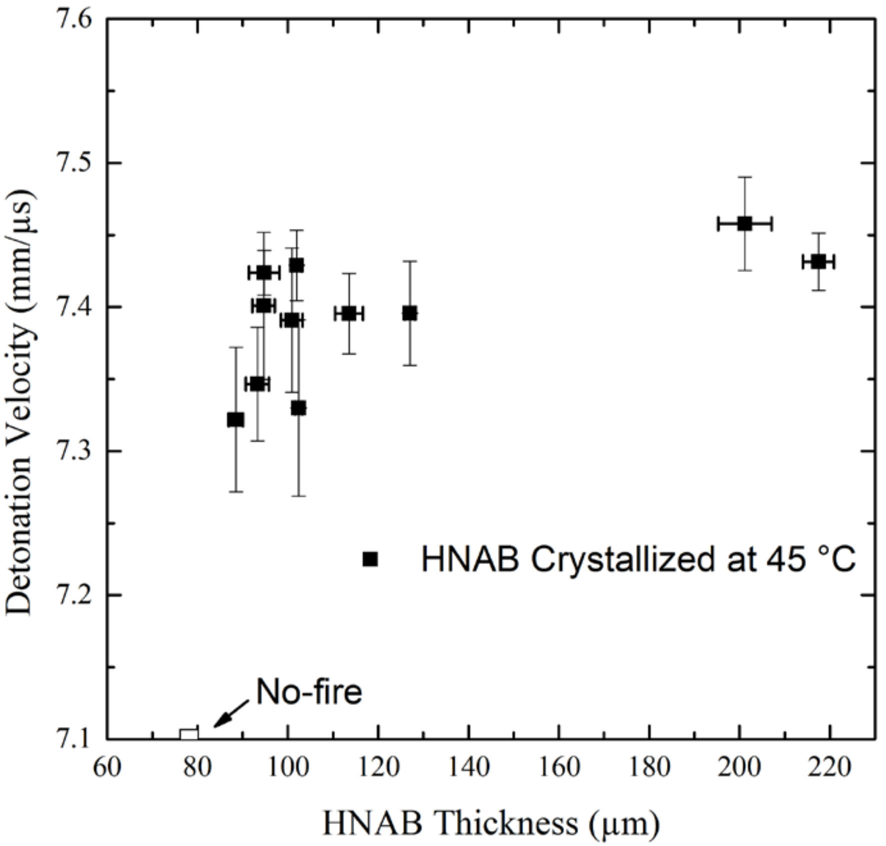
- Detonation velocity relatively consistent approaching failure
- Failure thickness = (thinnest Fire + thickest No-Fire)/2
- 64.7 µm failure thickness



Detonation velocity versus thickness. One failure point is included.

Failure thickness for HNAB crystallized at 45 °C

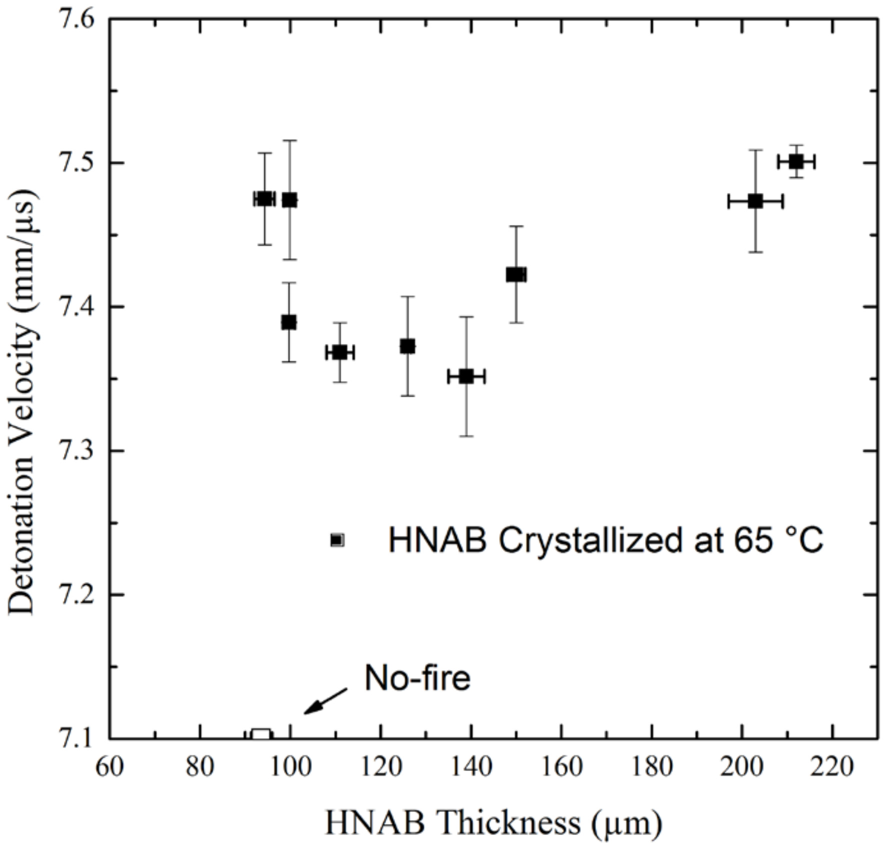
- Detonation velocity relatively consistent approaching failure
- Failure thickness = (thinnest Fire + thickest No-Fire)/2
- 83.4 µm failure thickness



Detonation velocity versus thickness. One failure point is included.

Failure thickness for HNAB crystallized at 65 °C

- Detonation velocity relatively consistent approaching failure
- Failure thickness = (thinnest Fire + thickest No-Fire)/2
- 94.0 µm failure thickness

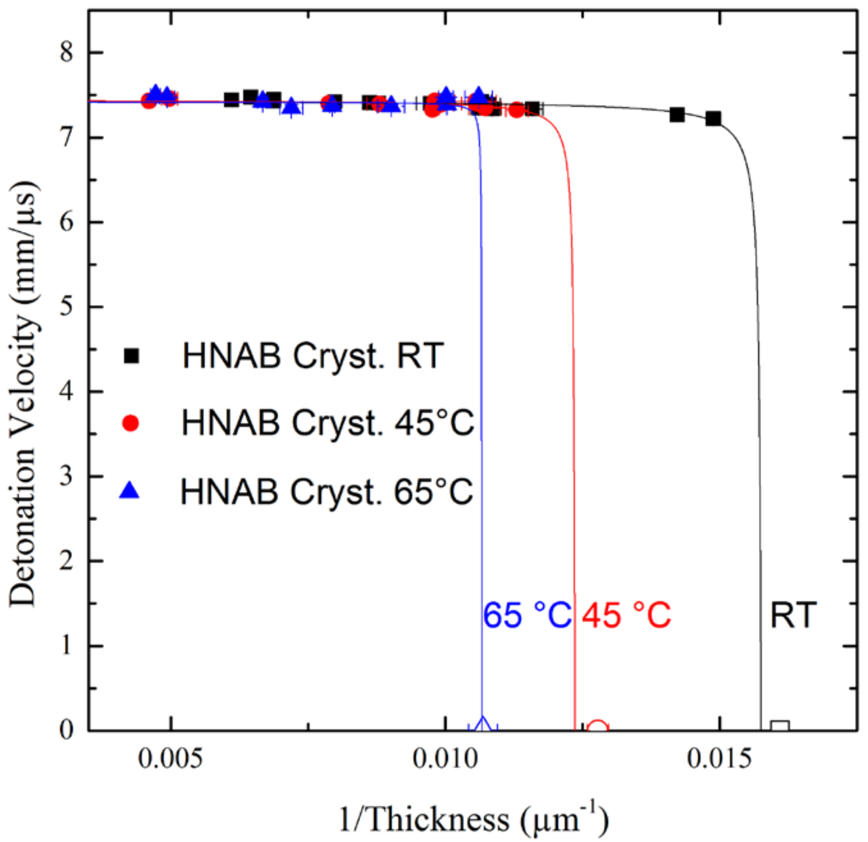


Detonation velocity versus thickness. One failure point is included.

Critical detonation thickness for room temperature, 45 °C, and 65 °C crystallized HNAB

$$D(R) = D(\infty)\left[1 - \frac{1}{t}\left(\frac{A}{1 - t_c\frac{1}{t}}\right)\right]$$

Cryst. Temperature	Detonation Velocity at Infinite Thickness, $D(\infty)$	Critical Thickness, t_c	Length Parameter, A
	mm/µs	µm	µm
Room T	7.424 ± 0.012	63.4 ± 1.3	0.120 ± 0.043
45 °C	7.435 ± 0.020	80.8 ± 2.6	0.109 ± 0.053
65 °C	7.417 ± 0.061	93.8 ± 0.2	0.025 ± 0.016

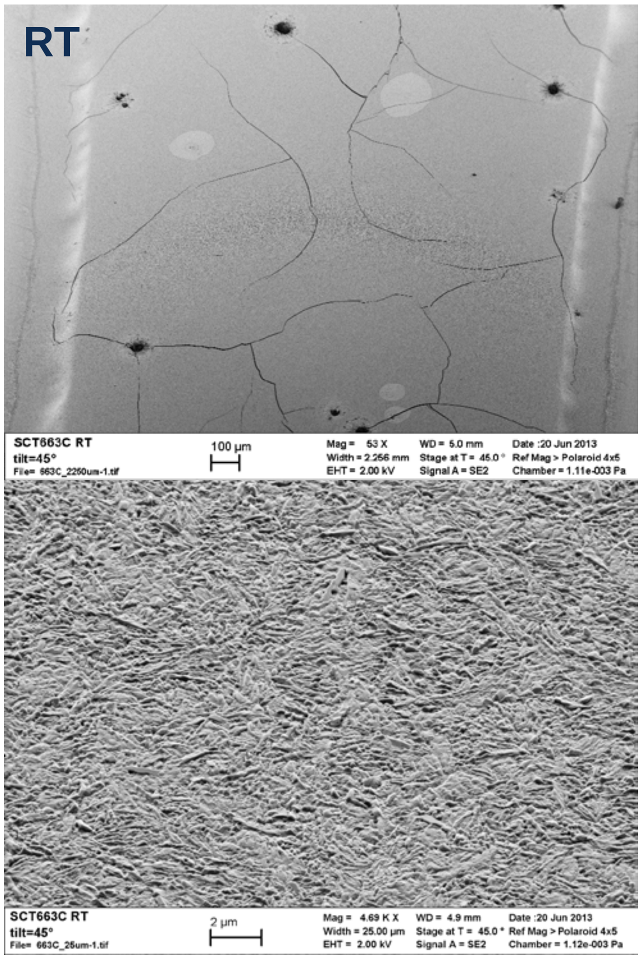


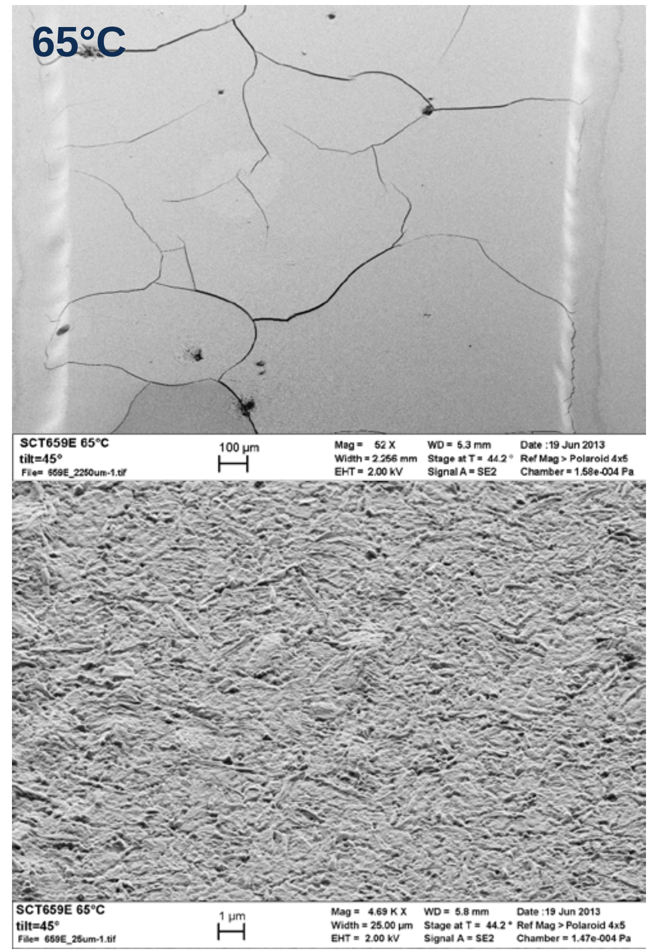
Detonation velocity versus 1/thickness.

Campbell, A.W. and Engelke, R., "The Diameter Effect in High-Density Heterogeneous Explosives," 6th Symposium (International) on Detonation, Coronado, CA, August 24-27, 1976, pp. 642-652.

Top surface microscopy of room temperature and 65 °C crystallized HNAB

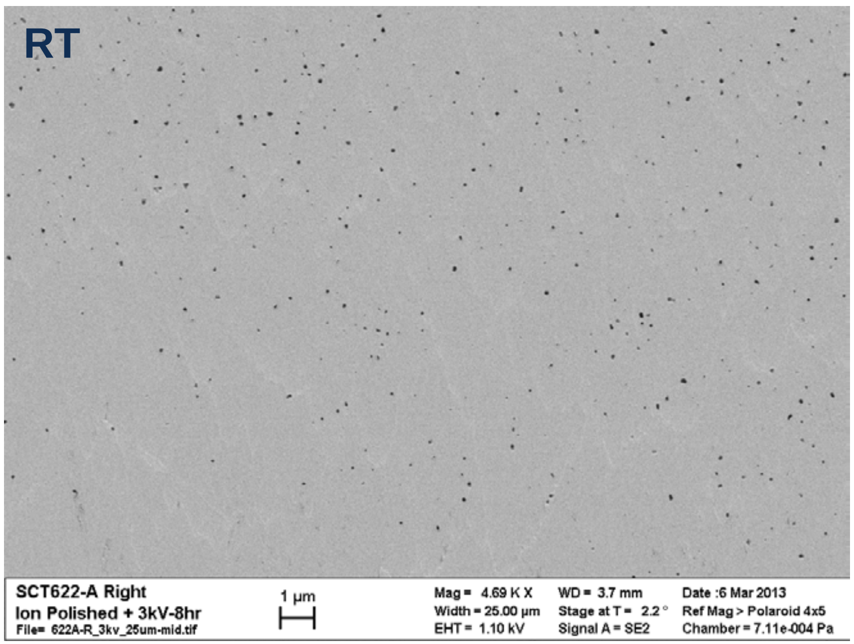
- Similar top surface appearance



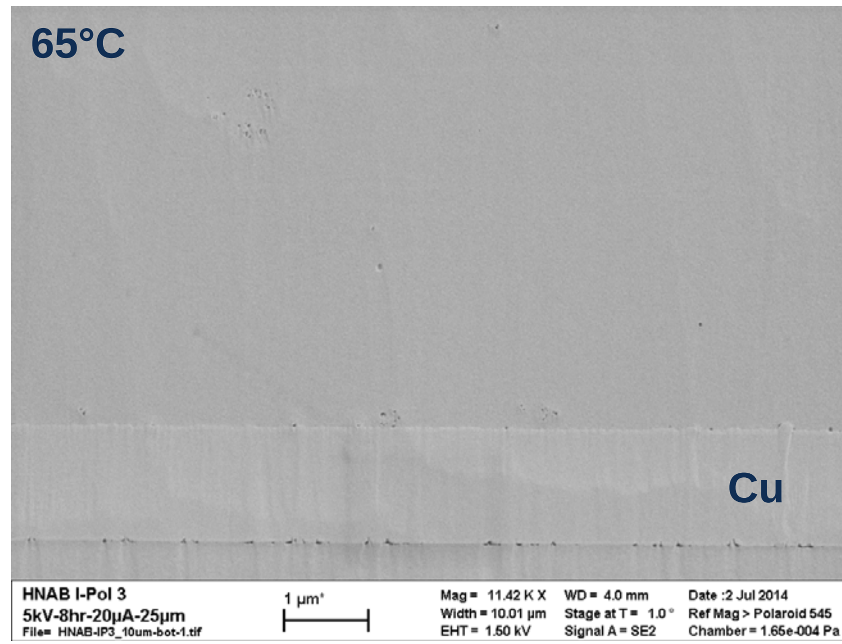


Ion-polished cross-sections of room temperature and 65 °C crystallized HNAB

- Pores (~ 100 nm) are distributed throughout room temperature crystallized HNAB, but not in 65 °C crystallized HNAB



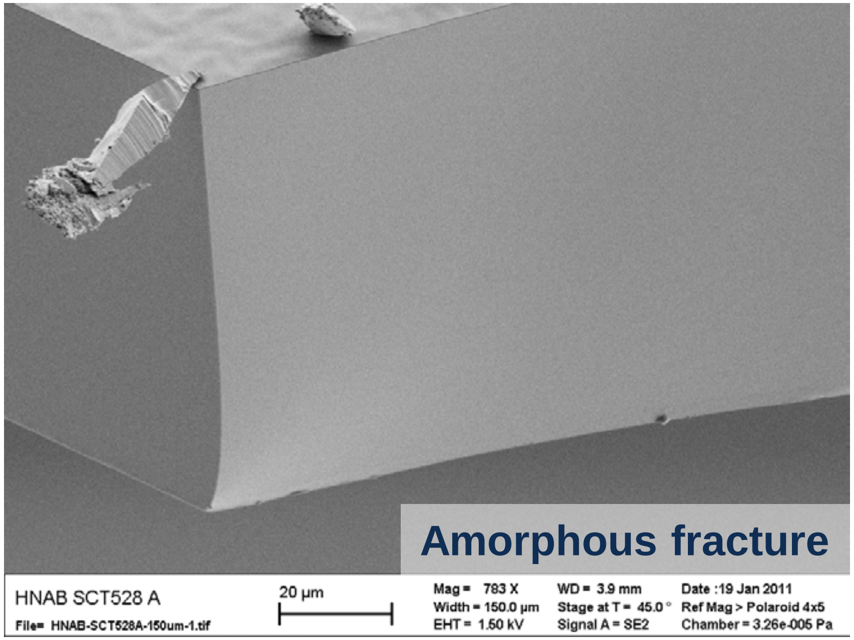
Scanning electron micrograph of ion polished room temperature crystallized HNAB. Field of view 25 µm.

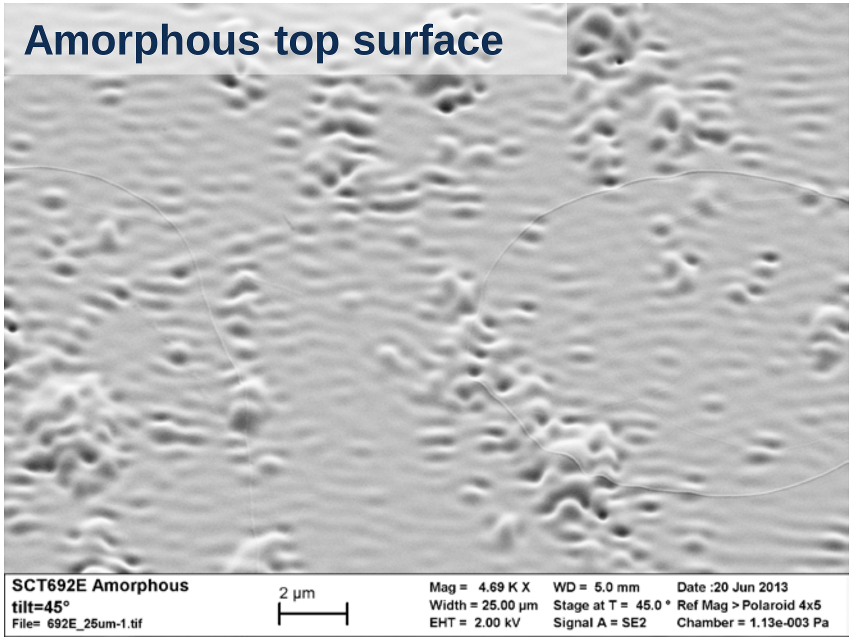


Scanning electron micrograph of ion polished 65 °C crystallized HNAB. Field of view 10 µm.

No detonation observed in amorphous HNAB or 85 °C crystallized HNAB

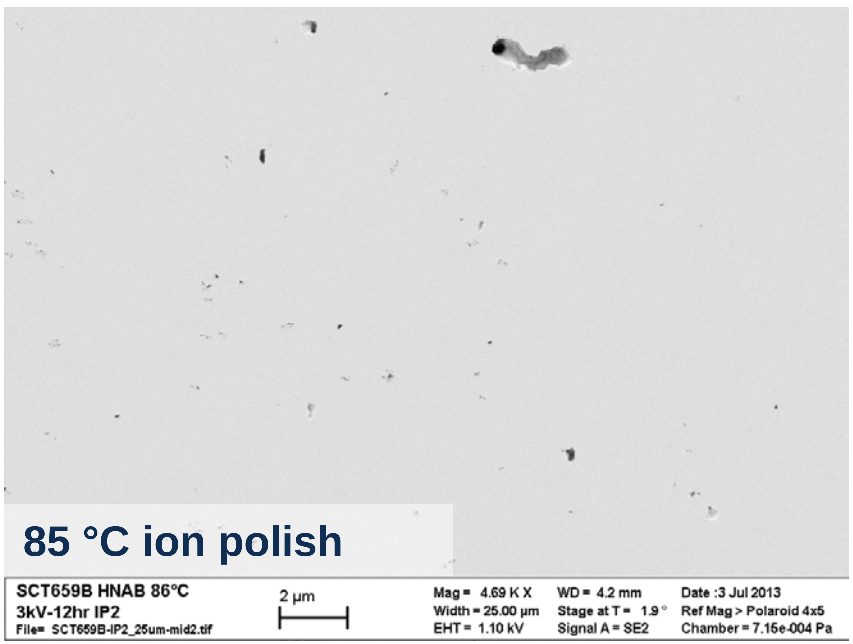
- In films up to ~ 200 µm thick
- No porosity in amorphous films

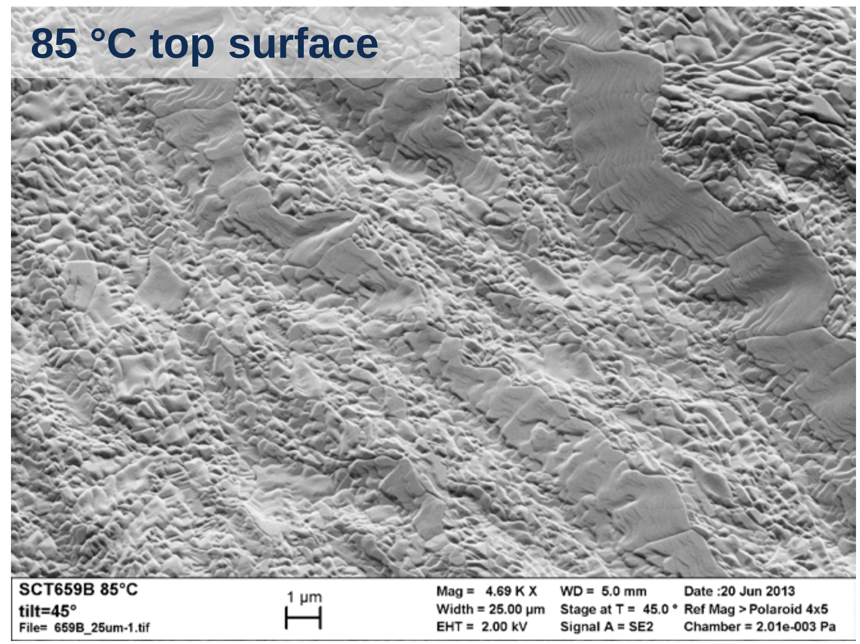




No detonation observed in amorphous HNAB or 85 °C crystallized HNAB

- In films up to ~ 200 µm thick
- Crystallization at 85 °C is above glass transition temperature



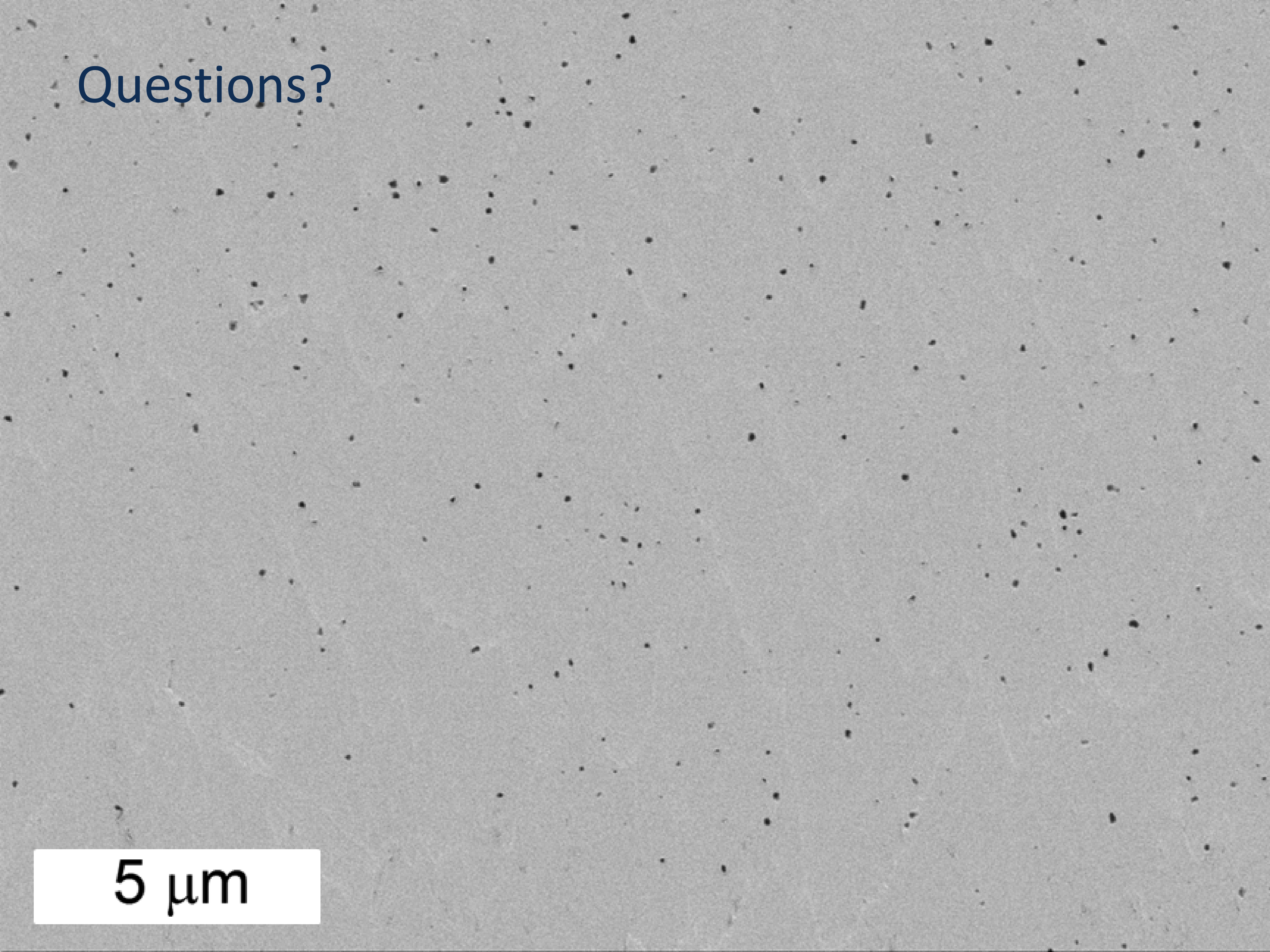


Conclusions

- Detonation in HNAB films depends on crystallization conditions and resulting porosity
  - Similar detonation velocities for films crystallized at room temperature, 45 °C and 65 °C
  - Smaller critical thickness with decreasing crystallization temperatures
  - Amorphous films lack porosity and do not detonate up to ~ 200 µm thick
  - Films crystallized at 85 °C (above T_g) do not detonate up to ~ 200 µm thick
- Acknowledgements

  - Michael P. Marquez, James Patrick Ball, Jill C. Miller, and M. Barry Ritchey
  - The Joint Department of Defense/Department of Energy Munitions Technology Development Program

Questions?



Backup slides

Surface profiler measurements

- Root mean squared surface roughness (R_q)
  - Amorphous films are smooth with a low R_q
  - Films crystallized below glass transition temperature (~70 °C) have crack networks and slightly higher R_q
  - Film crystallized above T_g higher R_q