Georgia Institute of Technology

Effects of Microstructure and Chemistry on Ignition Sensitivity of Polymer-bonded Explosives Under Shock Loading

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Georgia Institute of Technology

SSGF

STEWARDSHIP SCIENCE GRADUATE FELLOWSHIP

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Support by DTRA and SSGF

What are High Explosives?

- High Explosives (HEs) and their detonation properties have been an important field of study following WWII.

> - High explosives refer to explosive materials that detonate (i.e. supersonic shock fronts pass through material.

VS.

- Low explosives are subsonic → Flame front propagated by deflagration.

- Applications include:

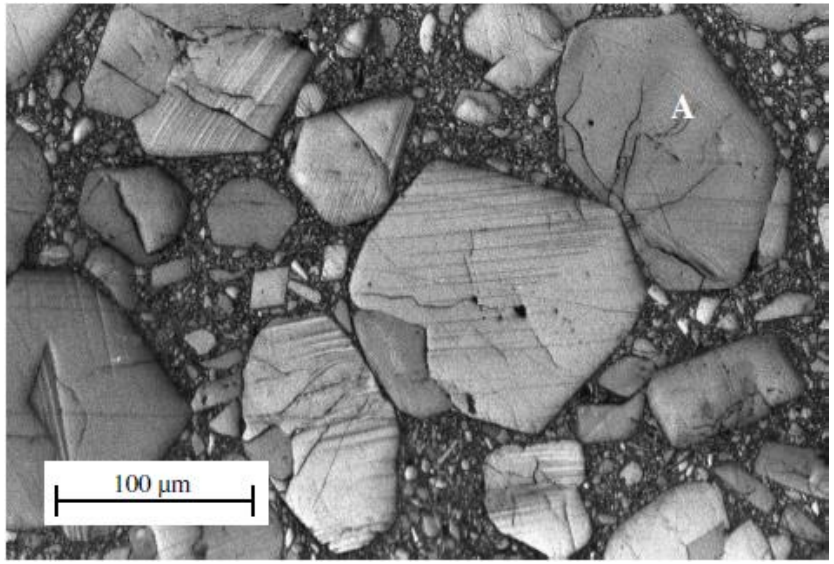
  - **Solid rocket propellants**
  - **Insensitive munitions**
  - **Tunnel construction**
  - **Demolition**
  - **Explosives in nuclear weapons**

↓

- Insensitive High Explosives
  - Palomares B-52 Plane Crash, 1966

Polymer Bonded Explosive (PBX)

- Primarily composed of 2 or 3 parts:
  - **Explosive Grains**
    - Brittle fracture under low pressure
    - Plastic deformation under high pressure
  - **Binder**
    - Bonding between constituents
    - Viscoelastic behavior
    - Low stiffness
  - **Additives**
    - Low sensitivity

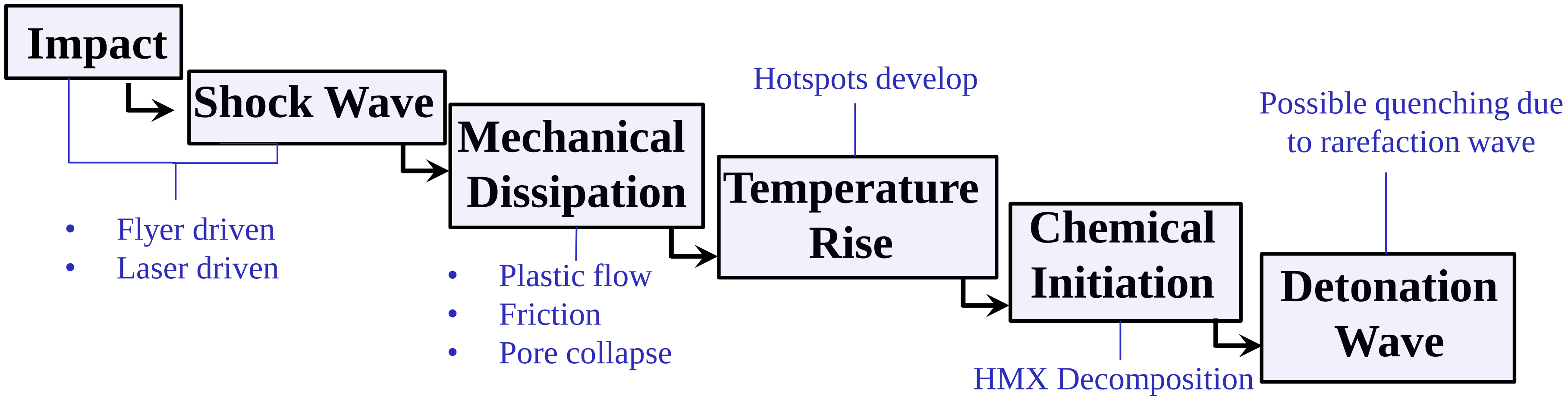


Rae *et al.*, 2002.

PBX constituents

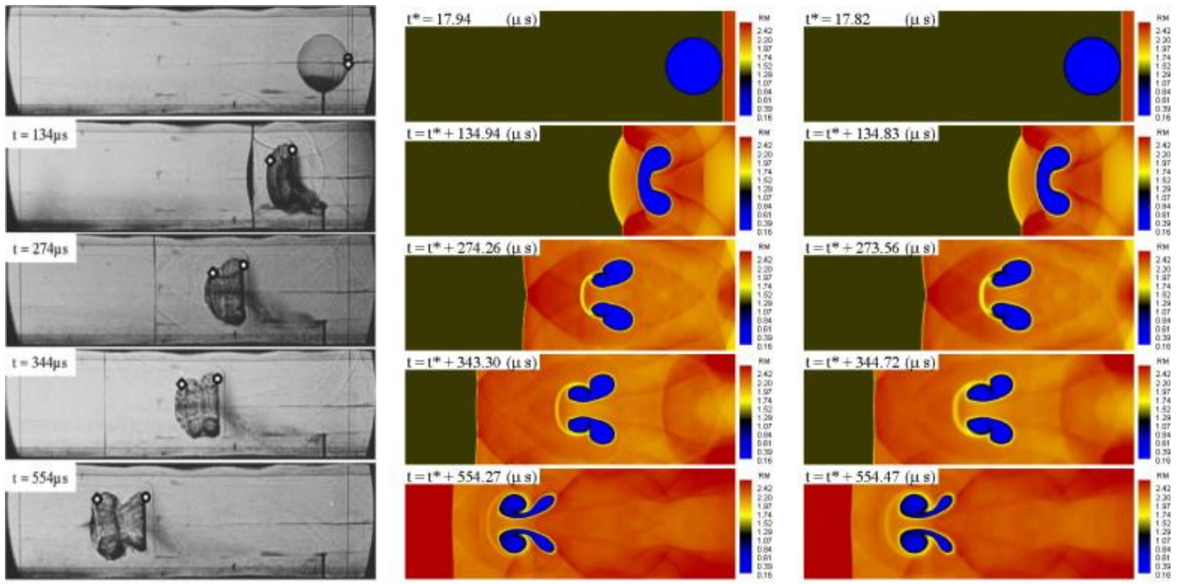
Explosive Grains	Binder Systems	Additives	
HMX	Estane	Aluminum	**PBX 9501**
RDX	Viton	Ammonium Perchlorate (AP)	
PETN	HTPB	Wax	
TATB	Kel-F 800	Inert Materials	
HNS	Polyurethane		

How do PBXs detonate?



Modeling shock with void in Fortran using Navier-stokes equations

Ha *et al.*, 2015.



(a) Experiments

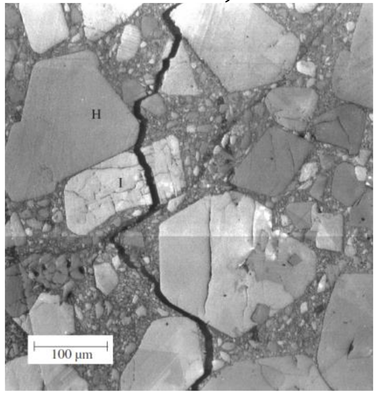
(b) Numerical results using six-equation model

(c) Numerical results using seven-equation model

Development of Hotspots

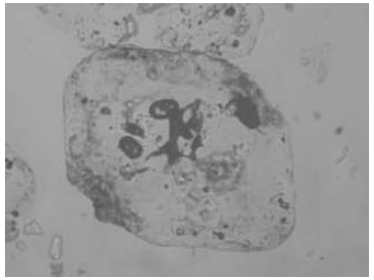
- ➢ Mechanical failure and interactions
  - – Inter-granular interactions
  - – Trans-granular fractures
  - – Debonding at grain-binder interface
- ➢ Defects in microstructures
  - – Imperfectly bonded interfaces
  - – Voids (Micropores)
- ➢ Shear bands (plastic flow)
  - – Localized heating along crystallographic slip planes

Rae et al., 2002.



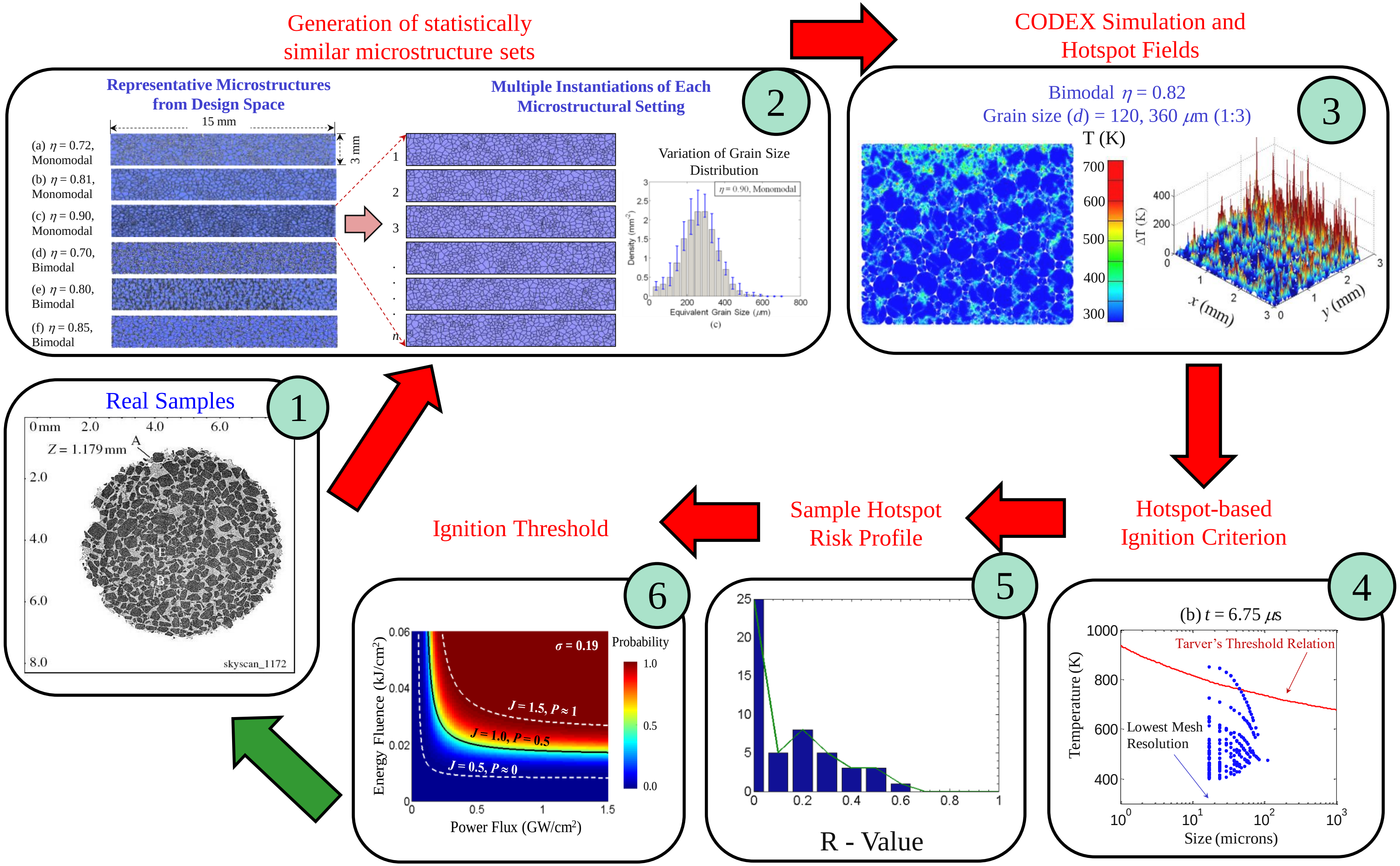
Fracture path

Czerski et al., 2007



Voids in RDX

CODEX Approach for Predicting Probabilistic Ignition Behavior of HEMs



Types of Microstructures Used

➢ Microstructural Effects

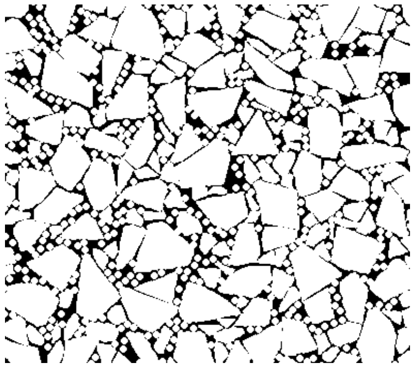
❖ Code is optimized to study the effects of mesoscale microstructural variables including the following:

- Microstructural heterogeneities
- Debonding
- Cracking
- Particulates
- Interfaces
- Various Types of dissipation

➢ Dissipation Mechanisms

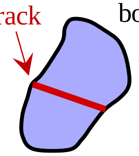
- Elasto-viscoplasticity
- Viscoelasticity
- Hyperelasticity
- Fracture
- Friction
- Heat transfer

PBX, Aluminized PBX



Transgranular crack

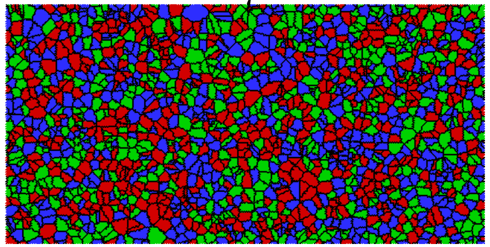
Intact grain boundary



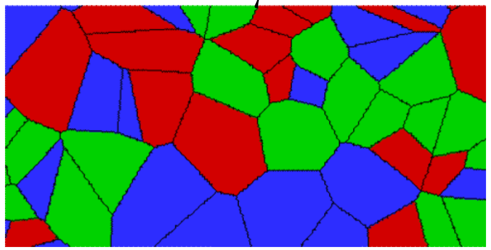
1 mm

Granular EM Beds

d = 35 μm

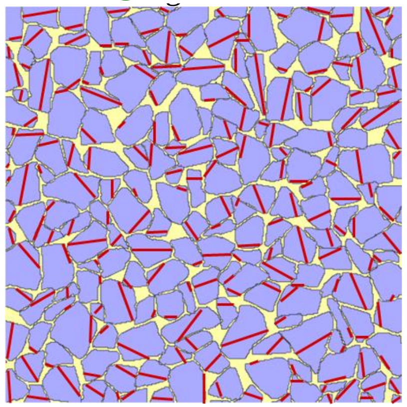


d = 220 μm



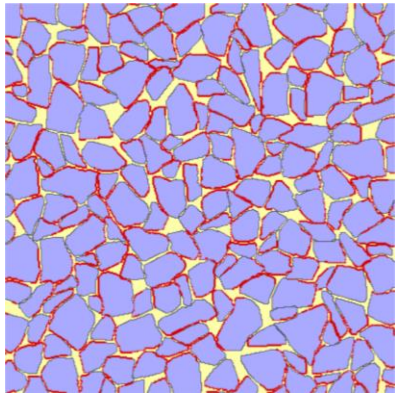
0.5 mm

Transgranular Cracks



1 mm

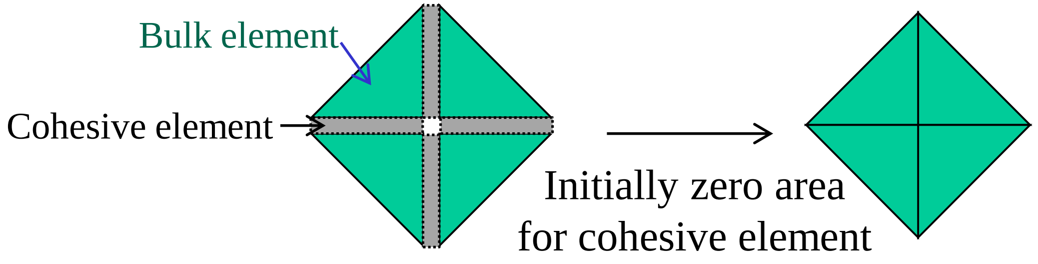
Debonding



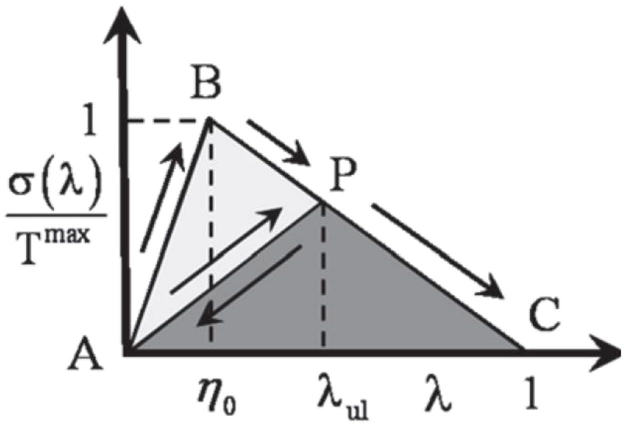
1 mm

CODEX Setup

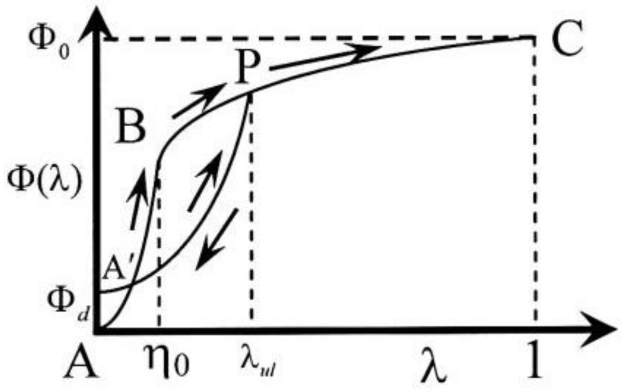
Cohesive FEM



Element configuration (bulk/cohesive element)

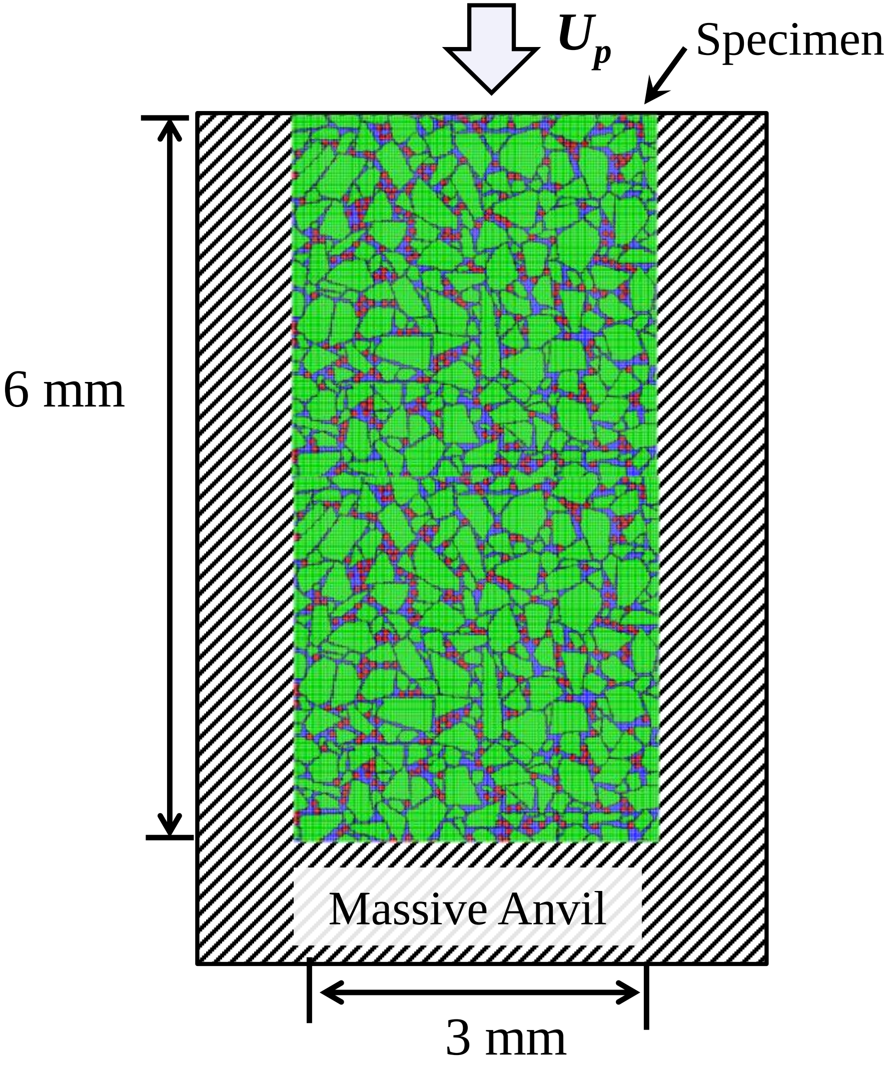


Bilinear traction-separation



Cohesive energy

Loading Conditions

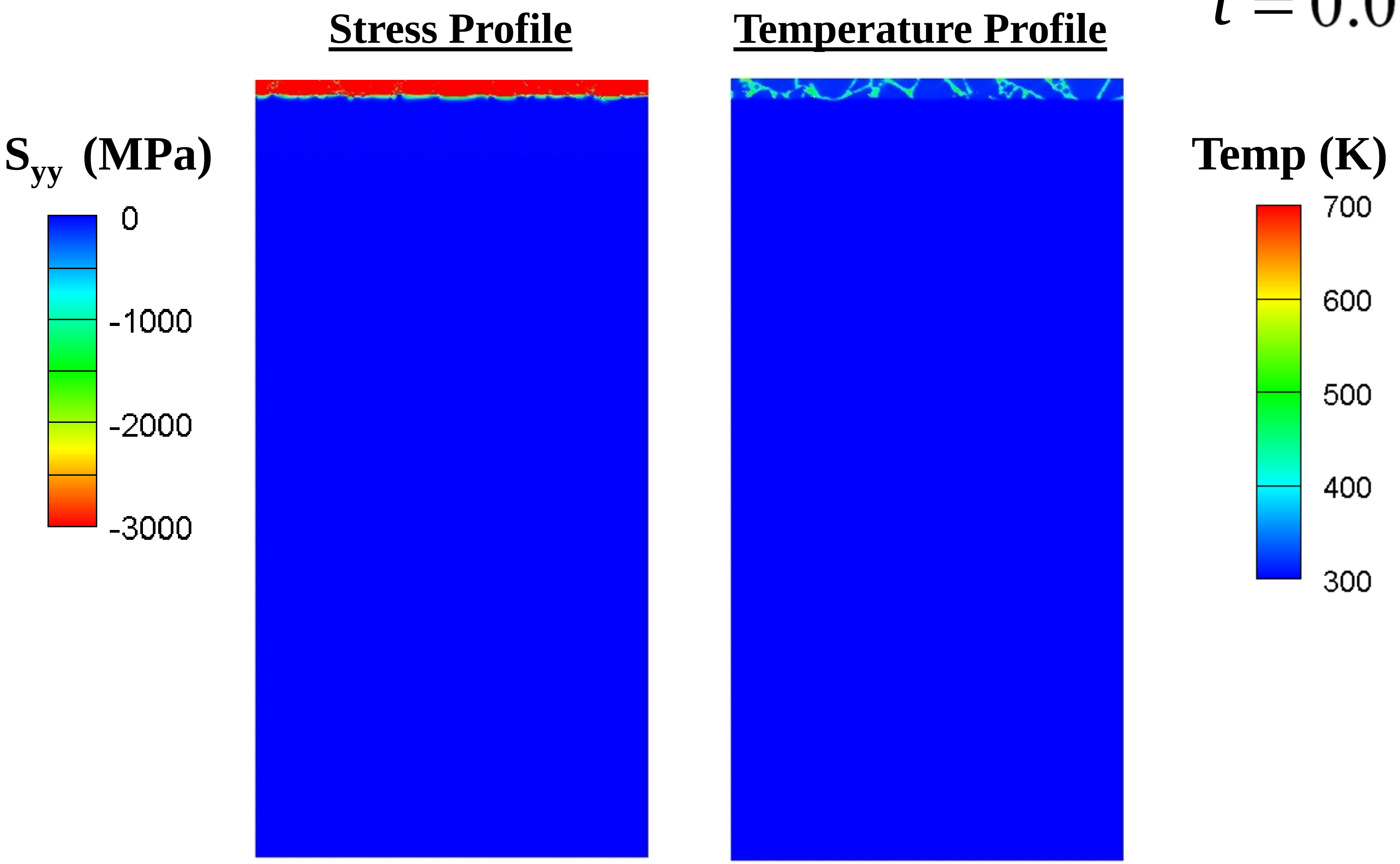


* *May and Tarver, AIP Conf. Proc.* **1195**, 275 (2009)

Temperature Field Evolution (high flyer velocity)

U_p = 500 m/s, Δt_{pulse} = 280 ns, 6% Al

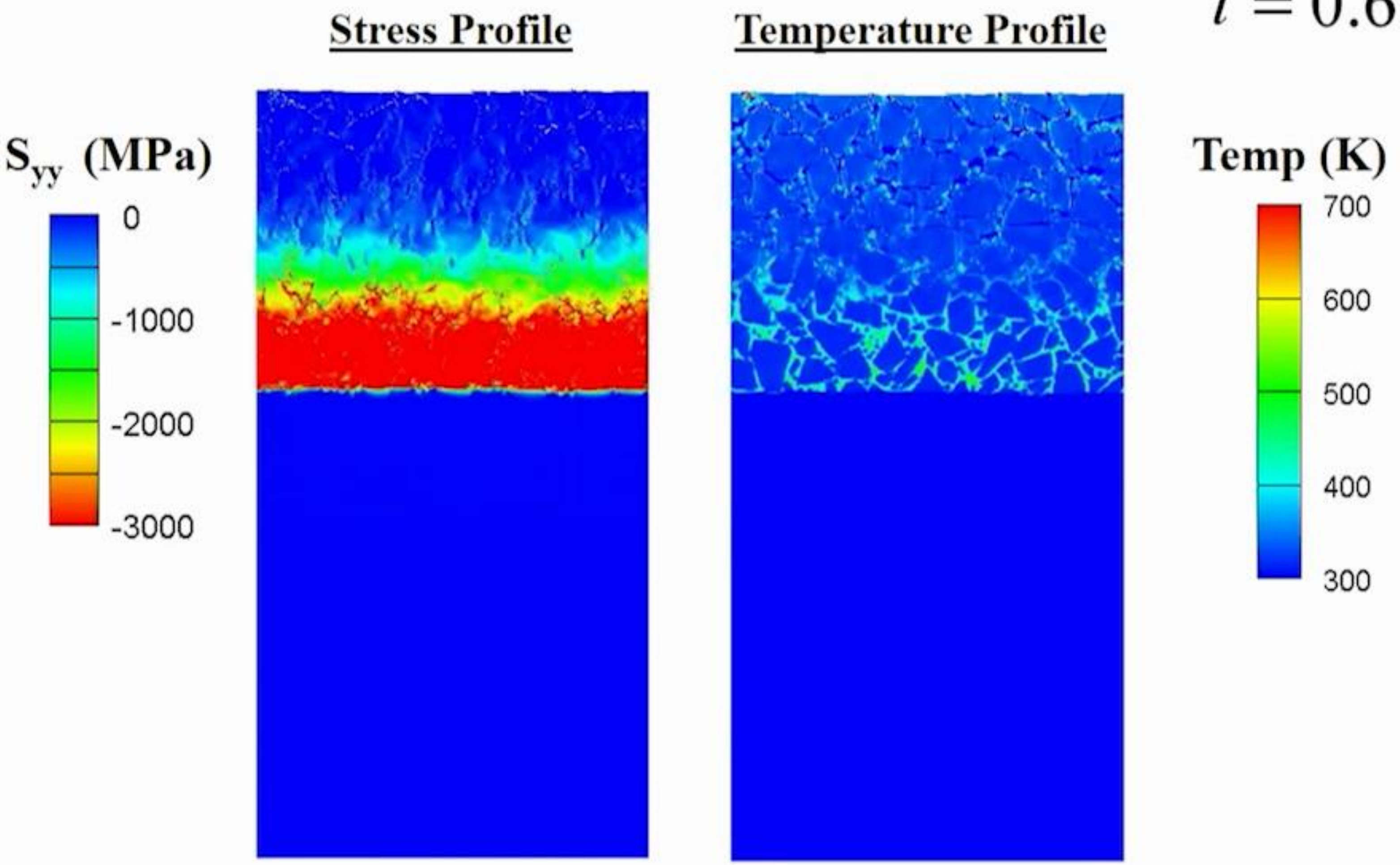
$t = 0.05\ \mu s$



Temperature Field Evolution (high flyer velocity)

U_p = 500 m/s, Δt_{pulse} = 280 ns, 6% Al

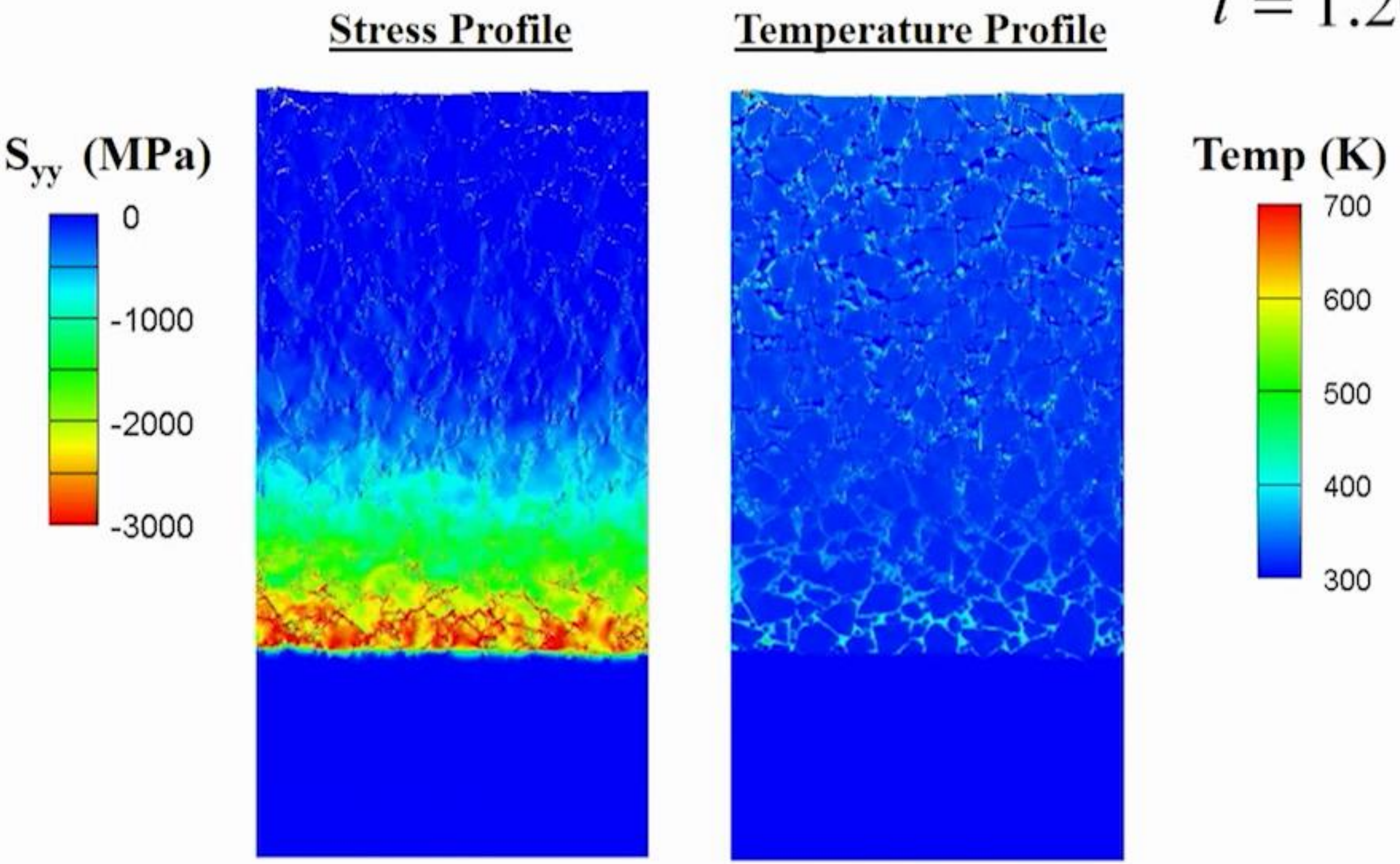
$t = 0.65\ \mu s$



Temperature Field Evolution (high flyer velocity)

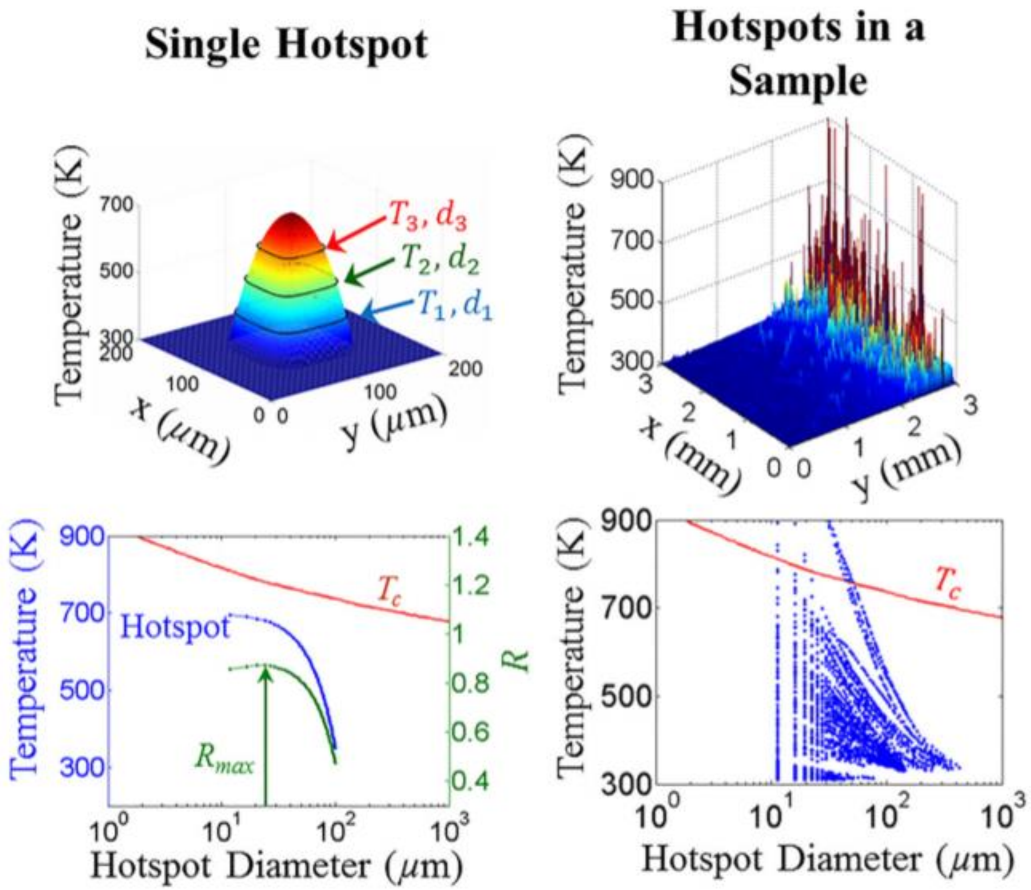
U_p = 500 m/s, Δt_{pulse} = 280 ns, 6% Al

t = 1.20 μs

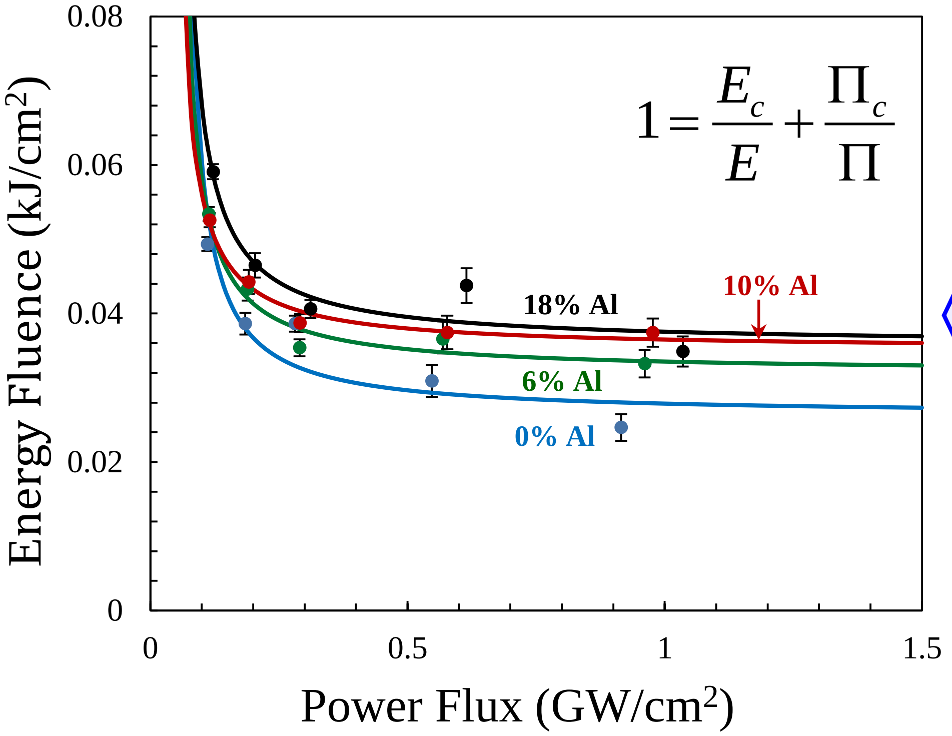


Determination of Ignition Likelihood via Hotspot Risk Analysis

- **Hotspot Ignition Risk Determinant (HIRD)**
  - Allows determination of criticality states of hotspots using a hotspot size-temperature ignition threshold
- **Ignition criterion for individual hotspots** (Barua *et al.* 2013)
  - $d\,(T) \geq d_c\,(T)$
- **Ignition criterion for microstructure**
  - If the density of hotspots exceeds 0.22 mm^{-2} at any point during the simulation. The sample is assumed to reach criticality.



Aluminized PBX Sensitivity Map

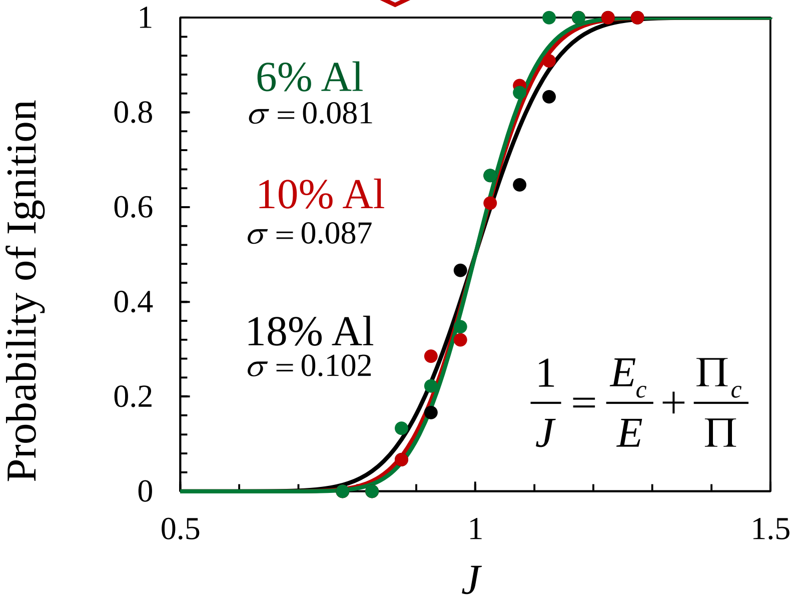


Aluminum Volume Fraction	E_c (kJ/cm²)	Π_c (GW/cm²)
0% Al	0.0263	0.0570
6% Al	0.0320	0.0452
10% Al	0.0351	0.0376
18% Al	0.0357	0.0479

- Higher volume fractions of aluminum are less sensitive.

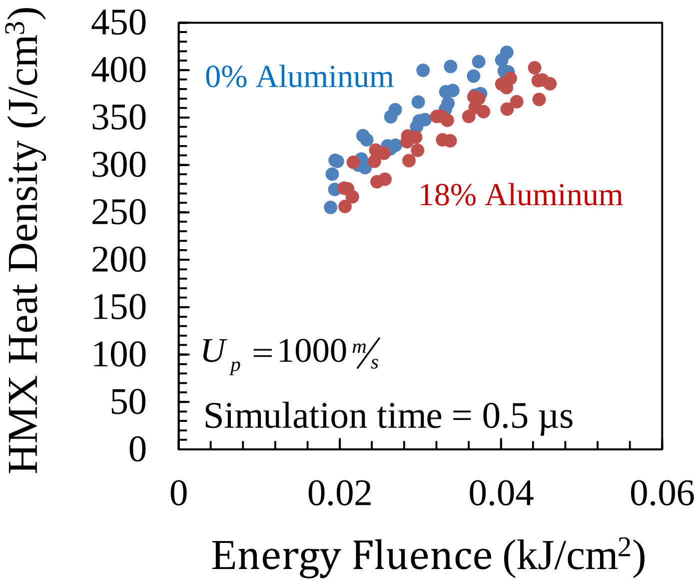
James Parameters are fit to 50% ignition threshold.

Entire probability distribution can be obtained via James parameters

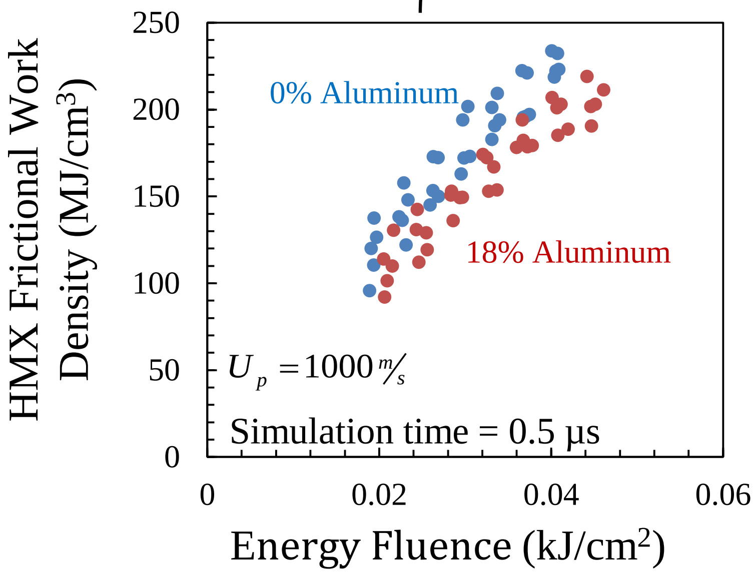


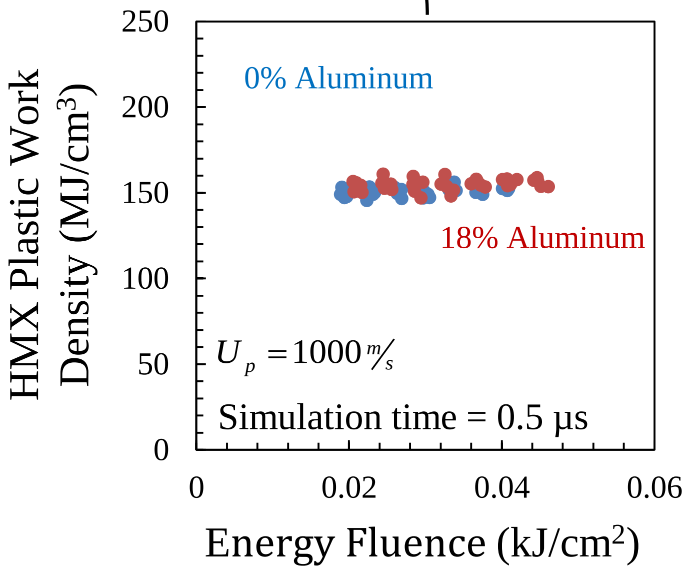
Heat Generation in HMX

- Friction generates more localized heat than plastic work
- Plastic work is responsible for the entire sample heating up, but not individual hotspots.



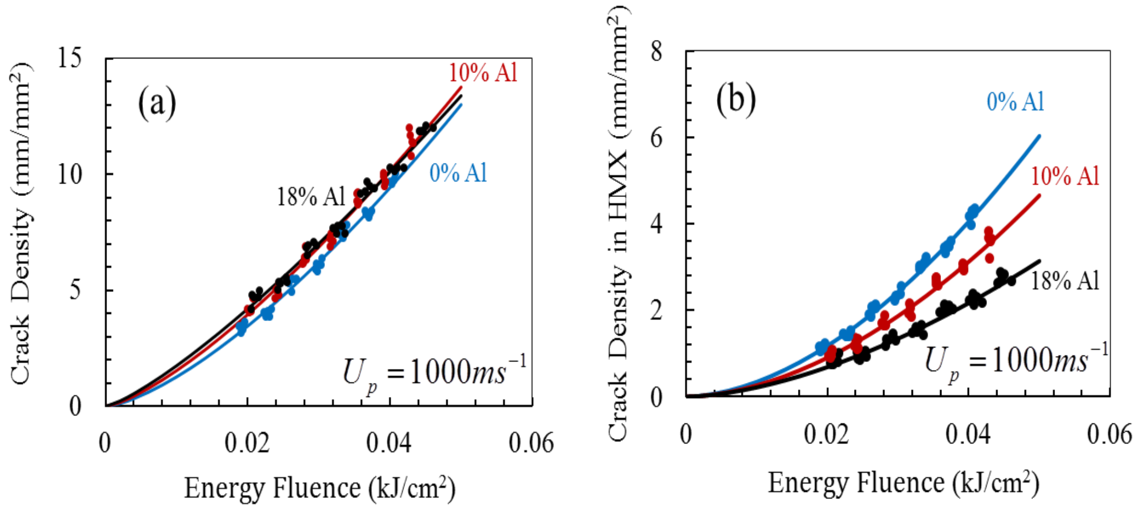
*Only areas above T = 350K were analyzed



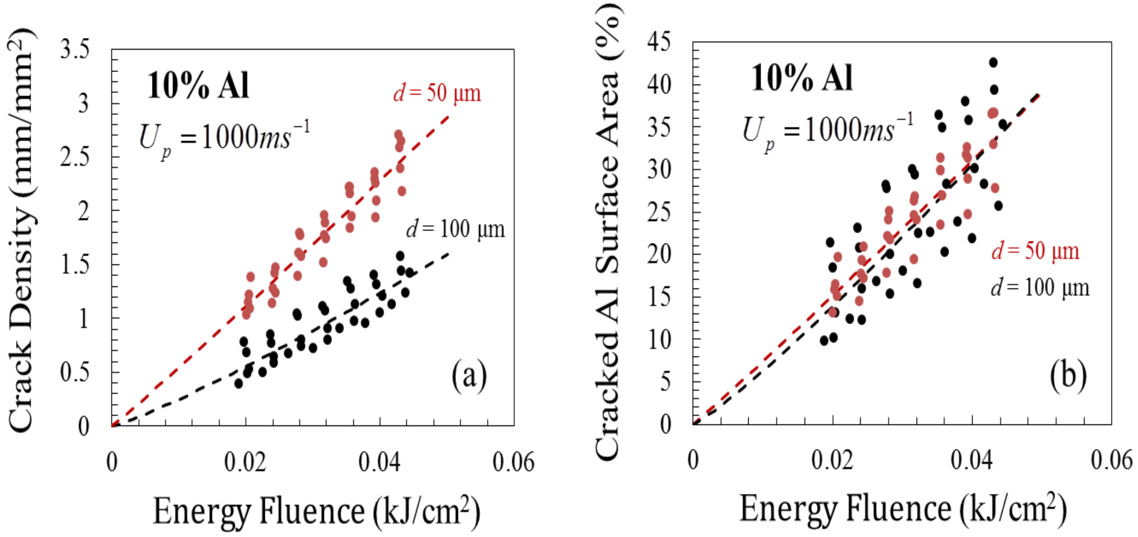


Effect of Aluminum on Fracture

- Total amount of cracks are similar between samples of varying Al concentration.
- When normalized to the relative amount of HMX, less HMX-associated cracks occur.



- Samples with smaller Al particles generate more cracks.
- When normalized to the relative surface area of aluminum, the proliferation of cracking is the same.



How do we consider detonation and void collapse?

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Sandia National Laboratories

CTH – An Eulerian Hydrocode

2D Microstructure-explicit/Voids-explicit Models (ME-VE-M)

➢ Statistically equivalent microstructure sample sets (SEMSS)

- Grains have monomodal size distribution with average size of 220 μm.
- SEMSS emulate experiments and allow evaluation of probabilistic/statistical nature of behavior.

➢ Microstructure effects

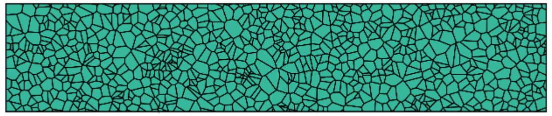
- Grains have random variations density ρ *
  - Hardin, Rimoli, and Zhou, *Thermochim Acta* **2014,** AIP Advances; 4, 097136 (2014)

$$\left\{ \begin{aligned} \rho_1 &= 2.473\,\mathrm{g/cm^3} \\ \rho_2 &= 1.902\,\mathrm{g/cm^3} \\ \rho_3 &= 1.331\,\mathrm{g/cm^3} \end{aligned} \right.$$

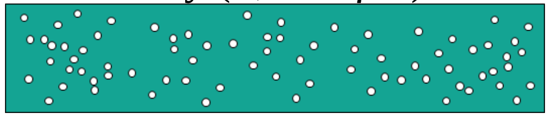
➢ ME-VE models (cases studied)

Homogeneous (H)

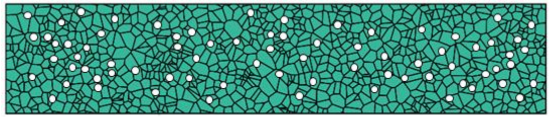
Microstructure Only (H)



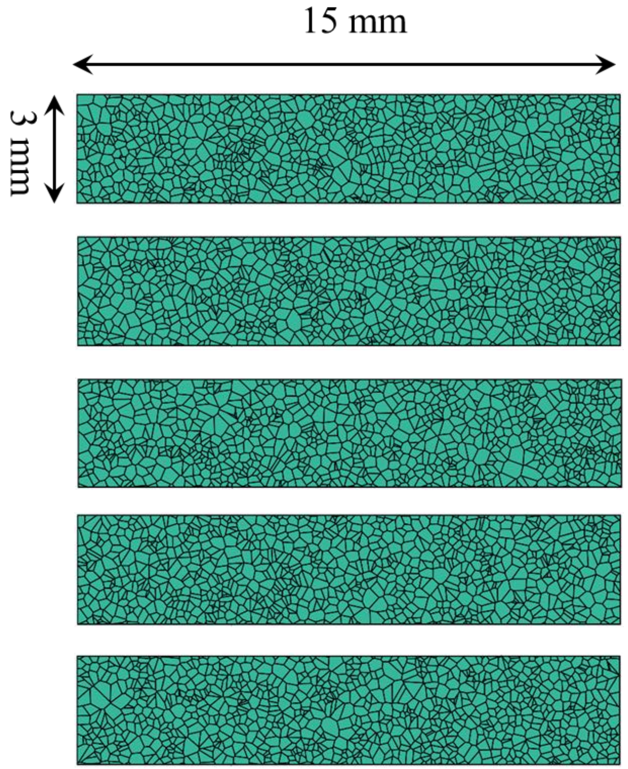
Voids Only (V, ~50 μm)

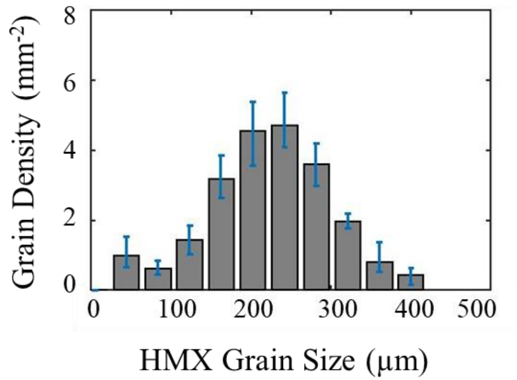


Voids + Microstructure (V+M)



SEMSS





Constitutive and Chemistry Models

Simplified Steinberg–Guinan–Lund (SGL) flow stress model

SGL: $Y = Y_A + Y_T(\dot{\varepsilon}, T),$ $\dot{\varepsilon}_p = \left[\frac{1}{C_1} \exp\left[\frac{2U_k}{T} \left(1 - \frac{Y_T}{Y_p} \right)^2 \right] + \frac{C_2}{Y_T} \right]^{-1}$

Mie-Grüneisen equation of state (MG-EOS)

MG-EOS: $P = \frac{\rho_0 C_0^2 (\eta - 1)\left[\eta - \frac{\Gamma_0}{2}(\eta - 1) \right]}{\left[\eta - s(\eta - 1) \right]^2} + \Gamma_0 E, \quad \eta = \frac{\rho}{\rho_0}, \quad E = \frac{1}{V_0} \int C_v dT$

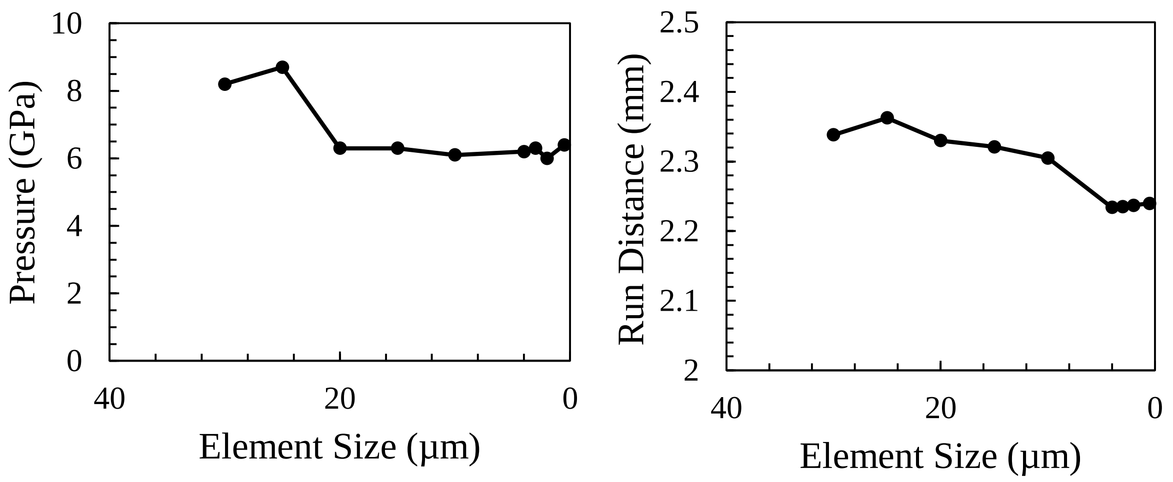
History Variable Reactive Burn (HVRB) chemistry model

HVRB: $\lambda = 1 - \left(1 - \phi^M / X \right)^X,$ $\phi = \tau_0^{-1} \int_0^t \left[(P - P_i)/P_R \right]^Z dt$

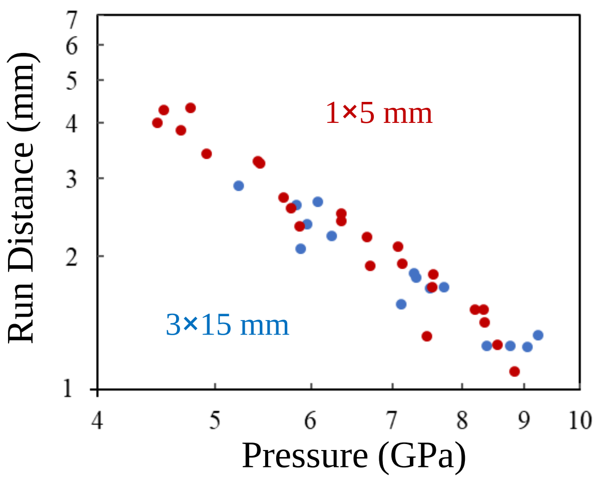
Computational Framework

- **Simulations performed using CTH – an Eularian-based hydrocode from SNL.**
- **Mesh convergence study performed.**
  - Samples have both microstructure and 50 μm voids.
  - Pressure converges more quickly than run distance (20 μm element vs. 5 μm element).
  - Final mesh resolution of 5 μm chosen.
- **A size comparison is needed to determine whether the samples are representative of a real size microstructure.**
  - **There is no significant discrepancy between 1×5 mm microstructures and 3×15 mm microstructures**

Mesh resolution

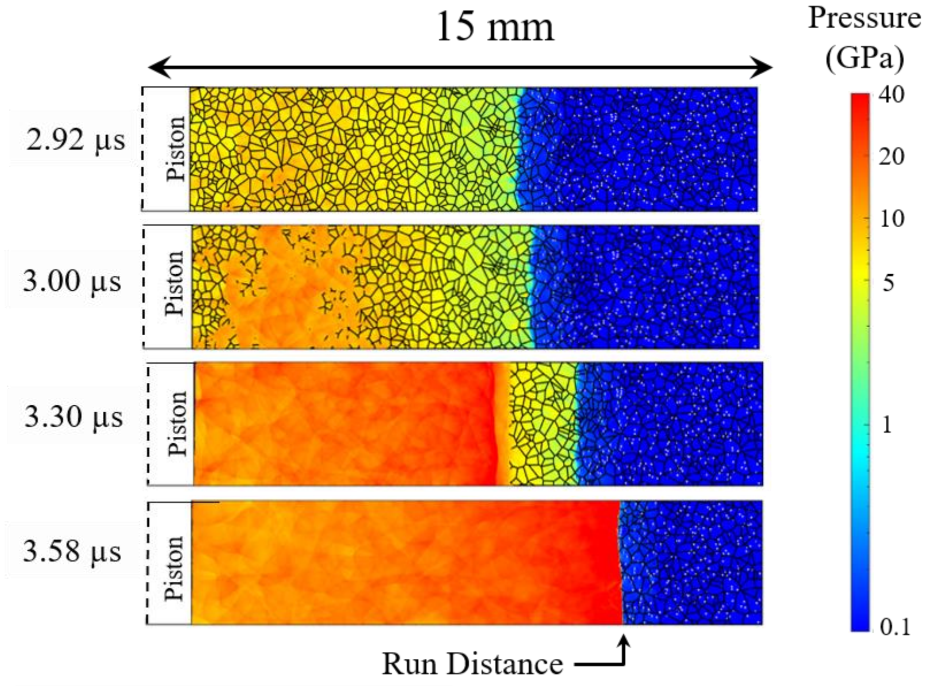


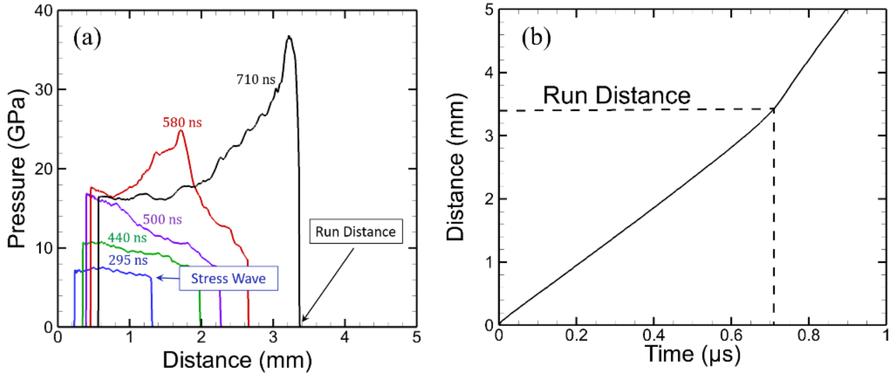
Sample size

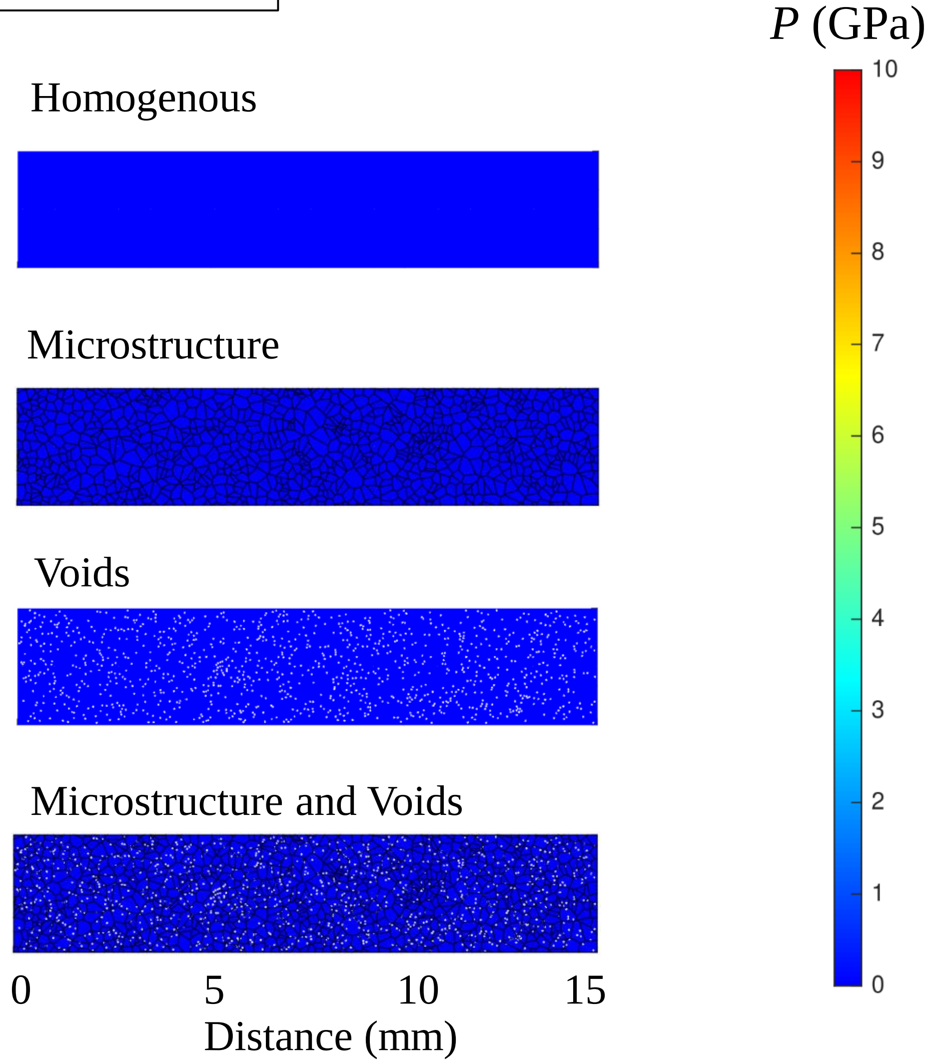


Shock to Detonation Transition (SDT)

U_p = 600 ms^{-1} (aluminum flyer), 30% MS variation







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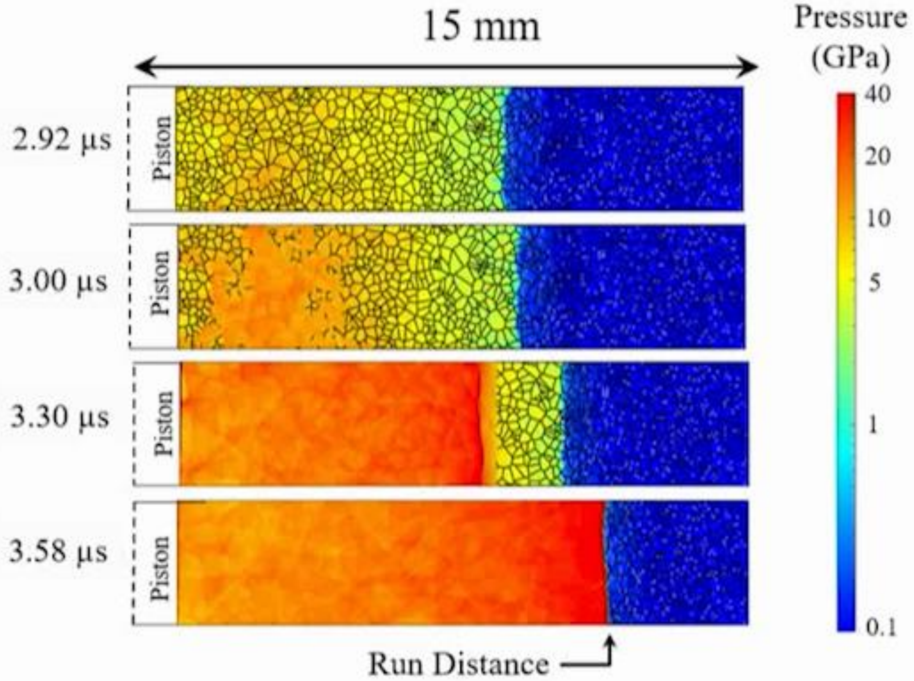

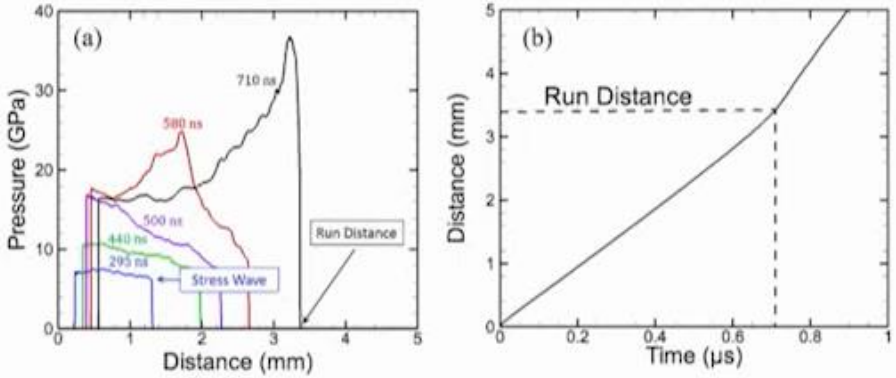

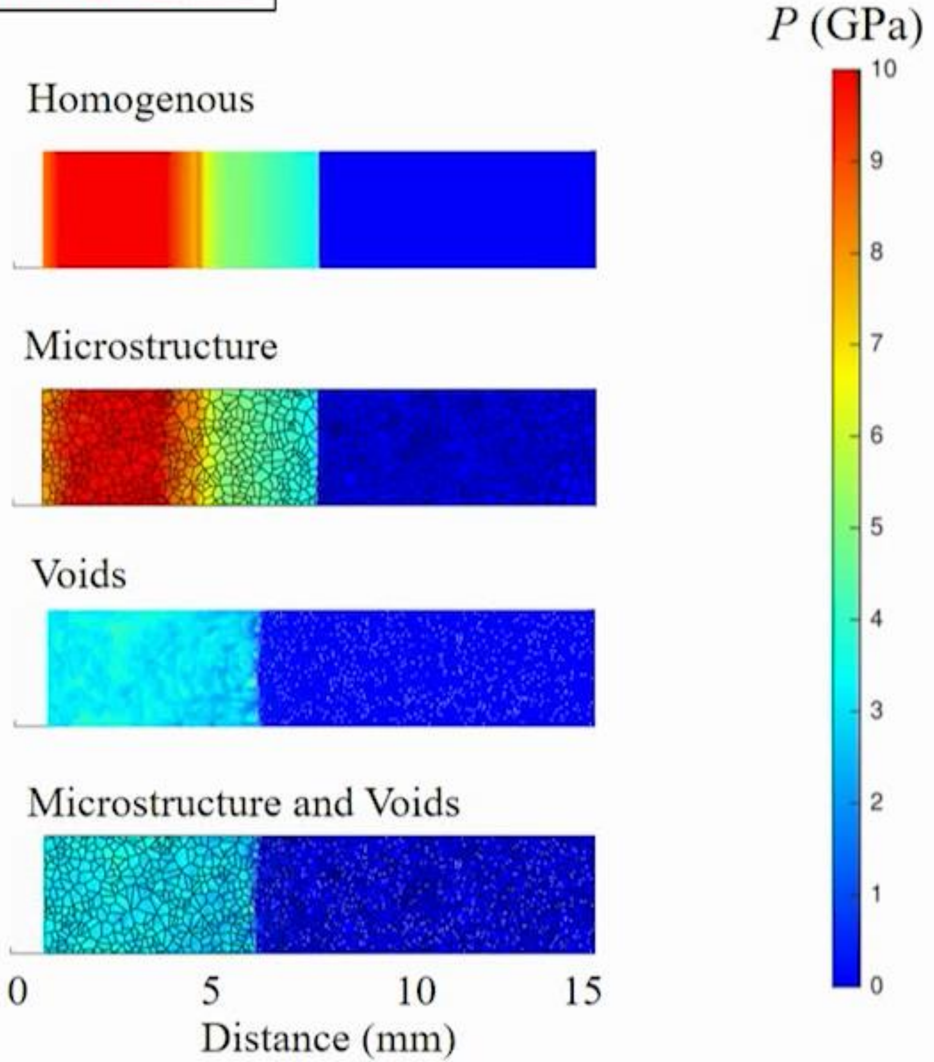

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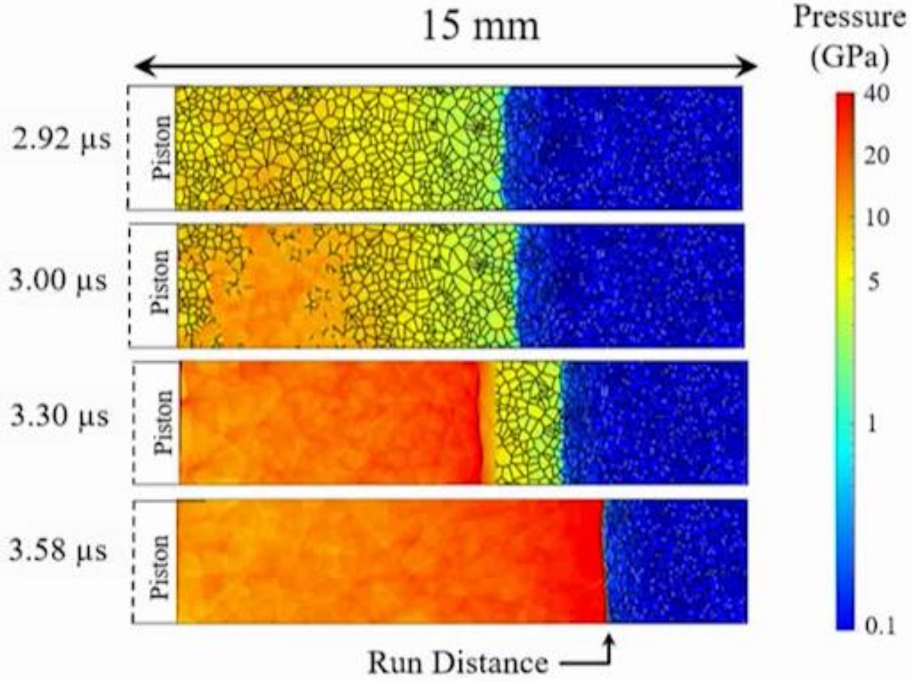
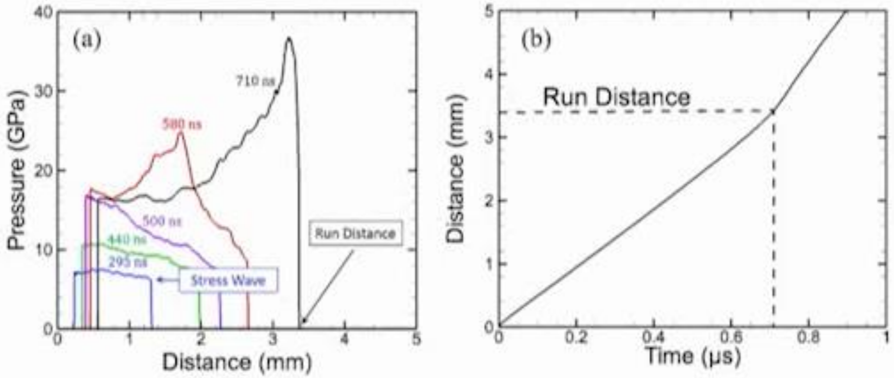
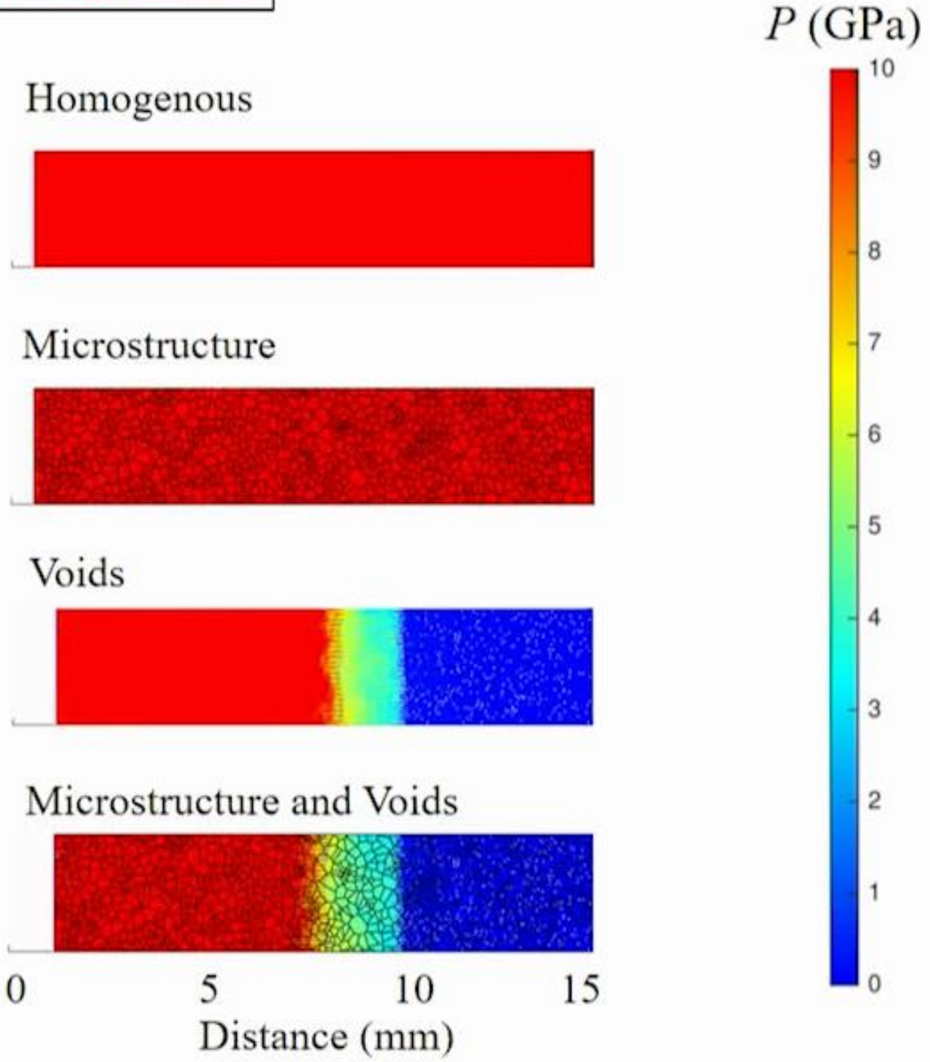
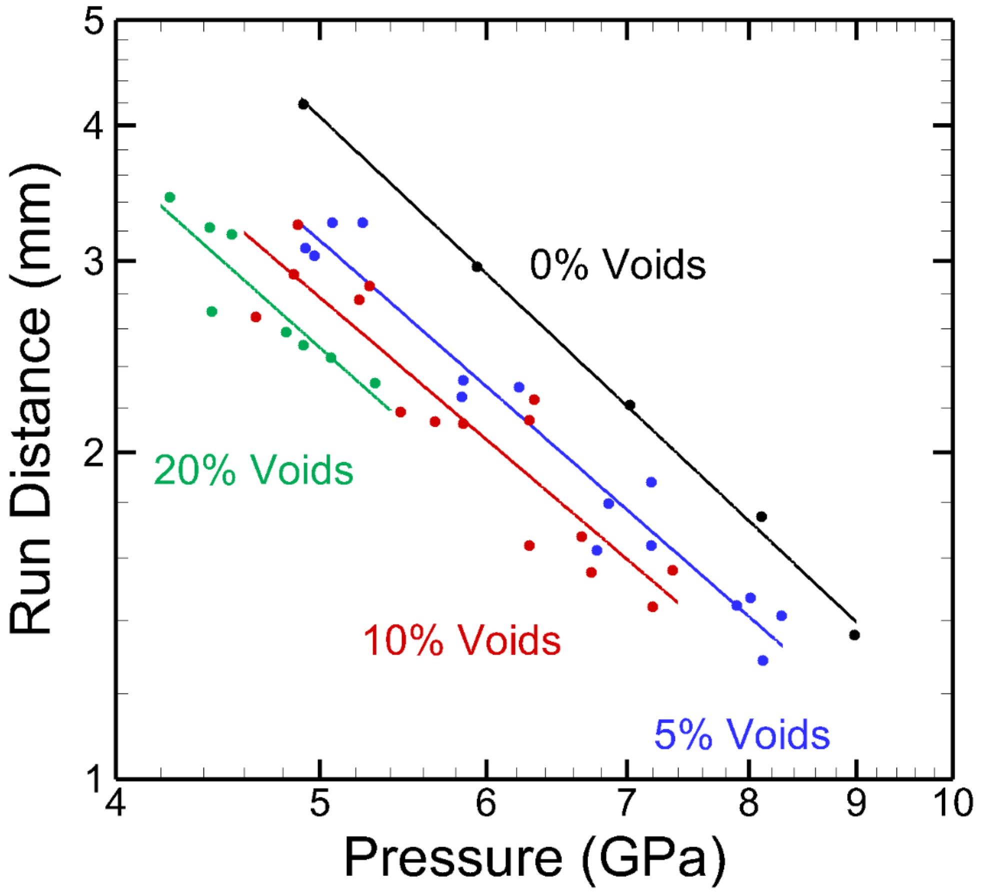
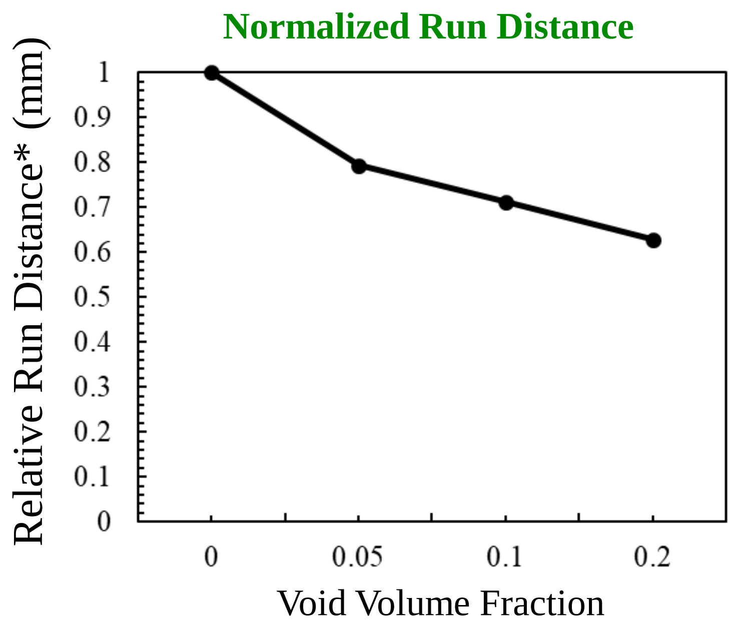






Effect of Voids on PP

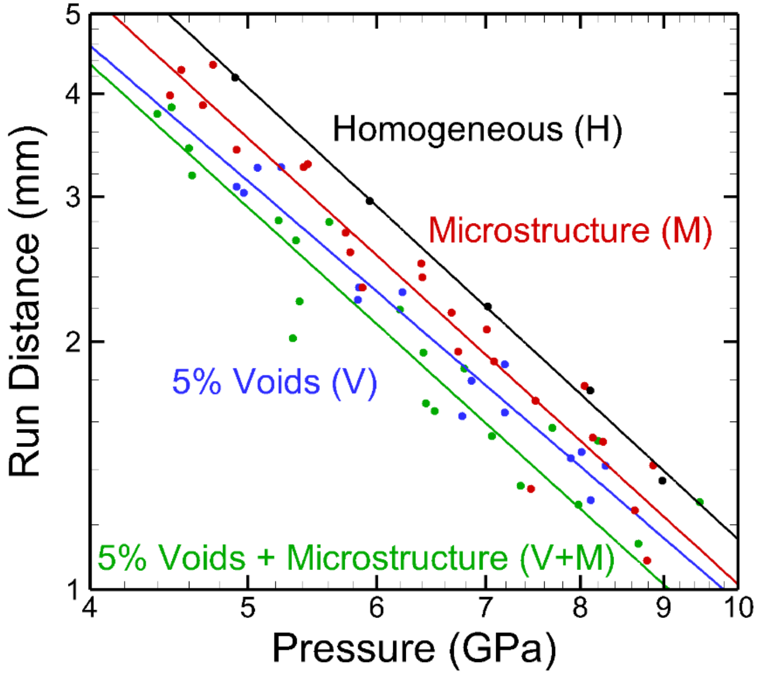


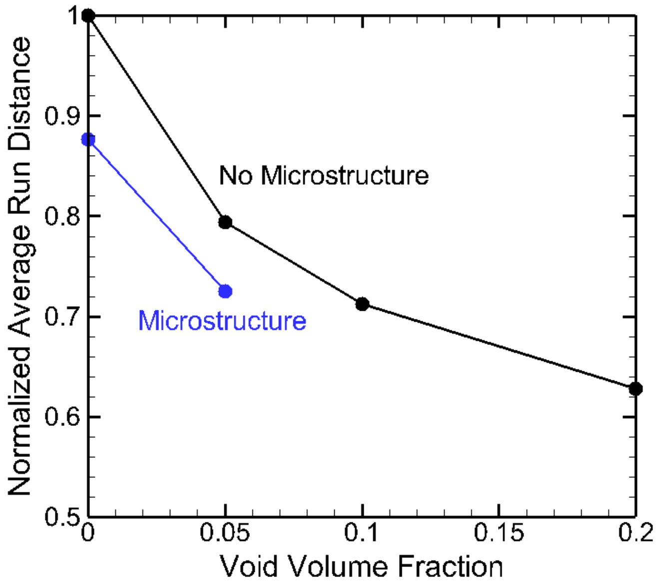


Void Volume Fraction	Average Decrease in Run Distance
5%	20.6%
10%	28.8%
20%	37.2%

Effects of Microstructure and Voids

Statistical Data Sets from SEMSS





HMX Sample Set	Average Decrease in Run Distance (compared to uniform)
Microstructure	12.2%
5% Voids	20.1%
5% Voids + Microstructure	27.5%

- Heterogeneities (microstructure, voids) enhance SDT sensitivity (shortens run distance or lowers PP line)
- Microstructure = constituent and morphological heterogeneities
- Voids = Geometric heterogeneities

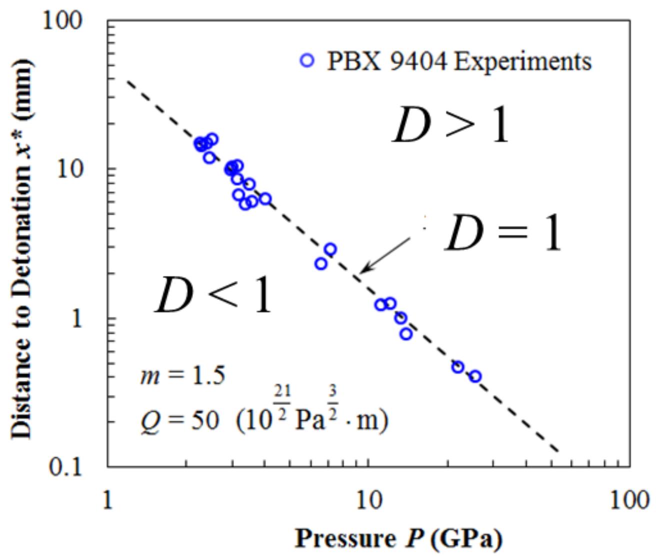
Probability formulation

- **Step 1: Pop plot fits are generally based on a power law relationship**

$$x^* = SP_s^{-m},$$

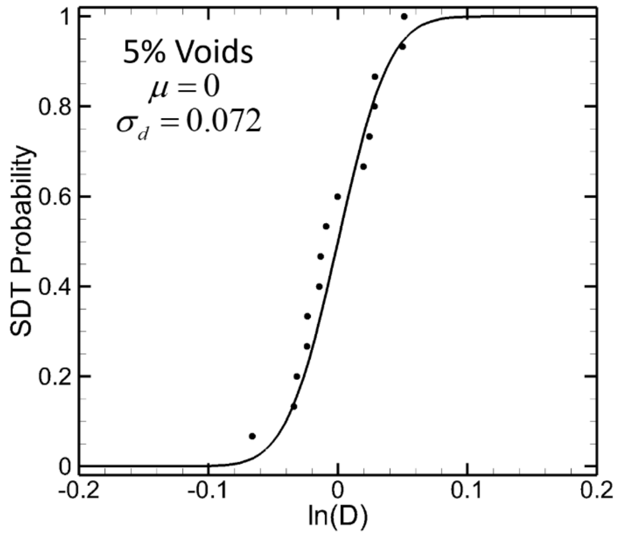
- **Step 2: Introduce a Non-dimensional Pop plot number (PPN) to gauge distance from PP**

$$D = \frac{(P_s - P_0)^m (x^* - x_0^*)}{S}.$$



- **Step 3: Create a probability formulation using a log-normal CDF**

$$\mathscr{P}(D) = \frac{1}{\sigma\sqrt{2\pi}} \int_0^D \frac{1}{x} \exp\left[\frac{-(\ln x - \mu)^2}{2\sigma_d^2}\right] dx,$$

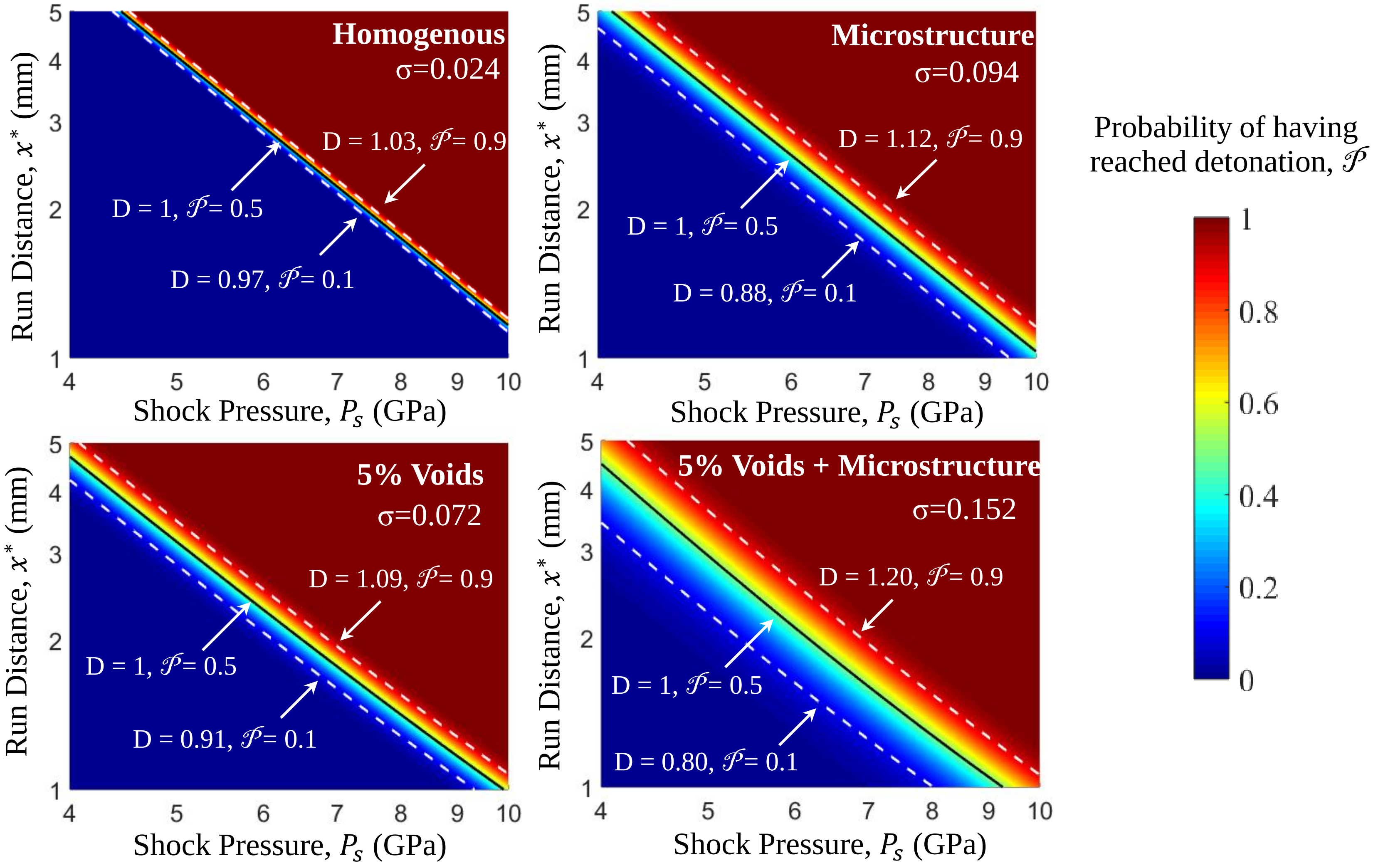


- **Step 4: Write in terms of physical parameters (shock pressure and run distance)**

$$\mathscr{P}(P_s, x^*) = \frac{1}{2} + \frac{1}{2}\mathrm{erf}\left[\frac{1}{\sqrt{2}\sigma_d}\left(\ln\left((P_s - P_0)^m\right) + \ln\left(x^* - x_0^*\right) - \ln S\right)\right].$$

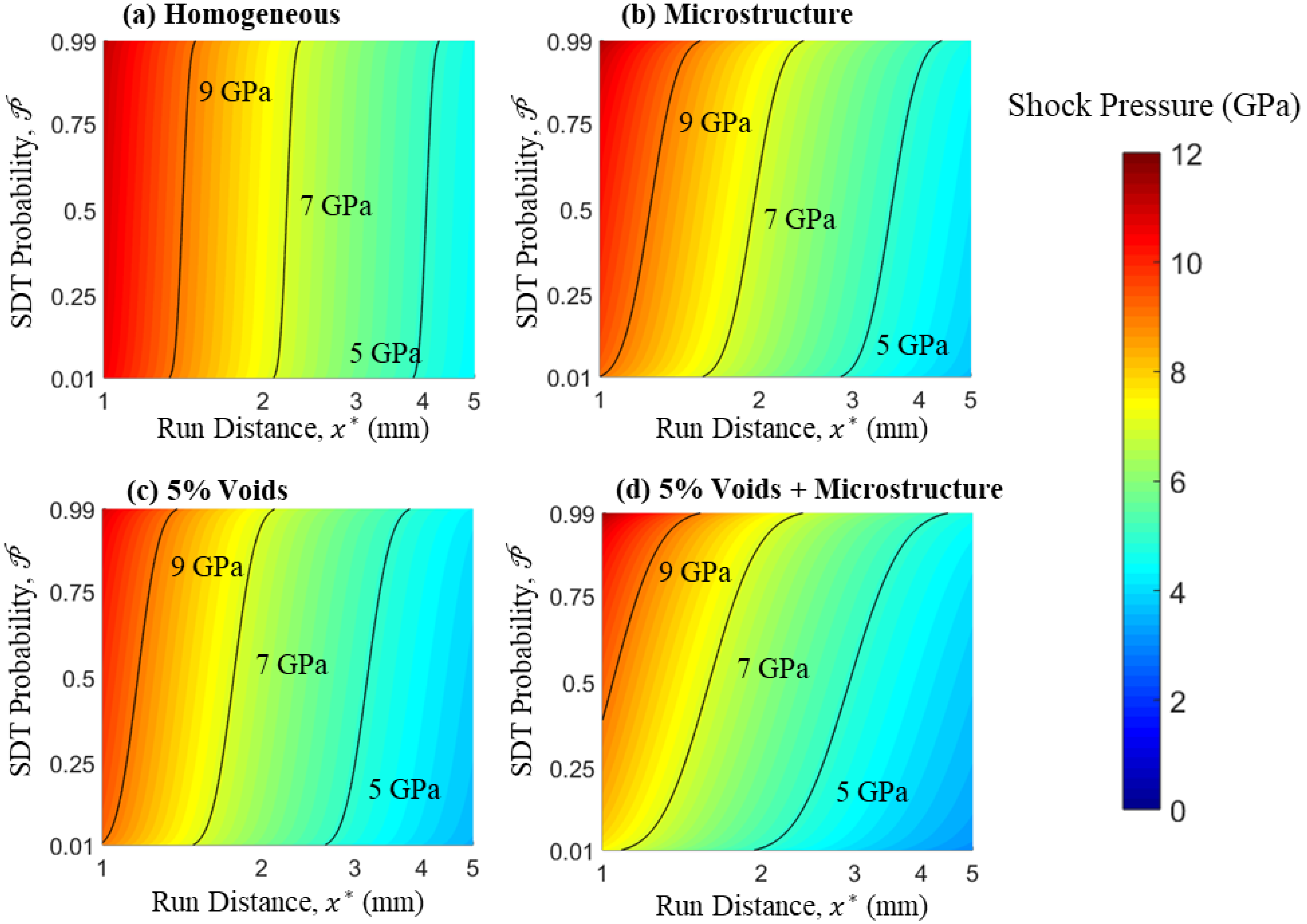
2D Probability Map

$$\mathscr{P}(P_s, x^*) = \frac{1}{2} + \frac{1}{2}\operatorname{erf}\left[\frac{1}{\sqrt{2}\sigma_d}\left(\ln\left(\left(P_s - P_0\right)^m\right) + \ln\left(x^* - x_0^*\right) - \ln S\right)\right].$$



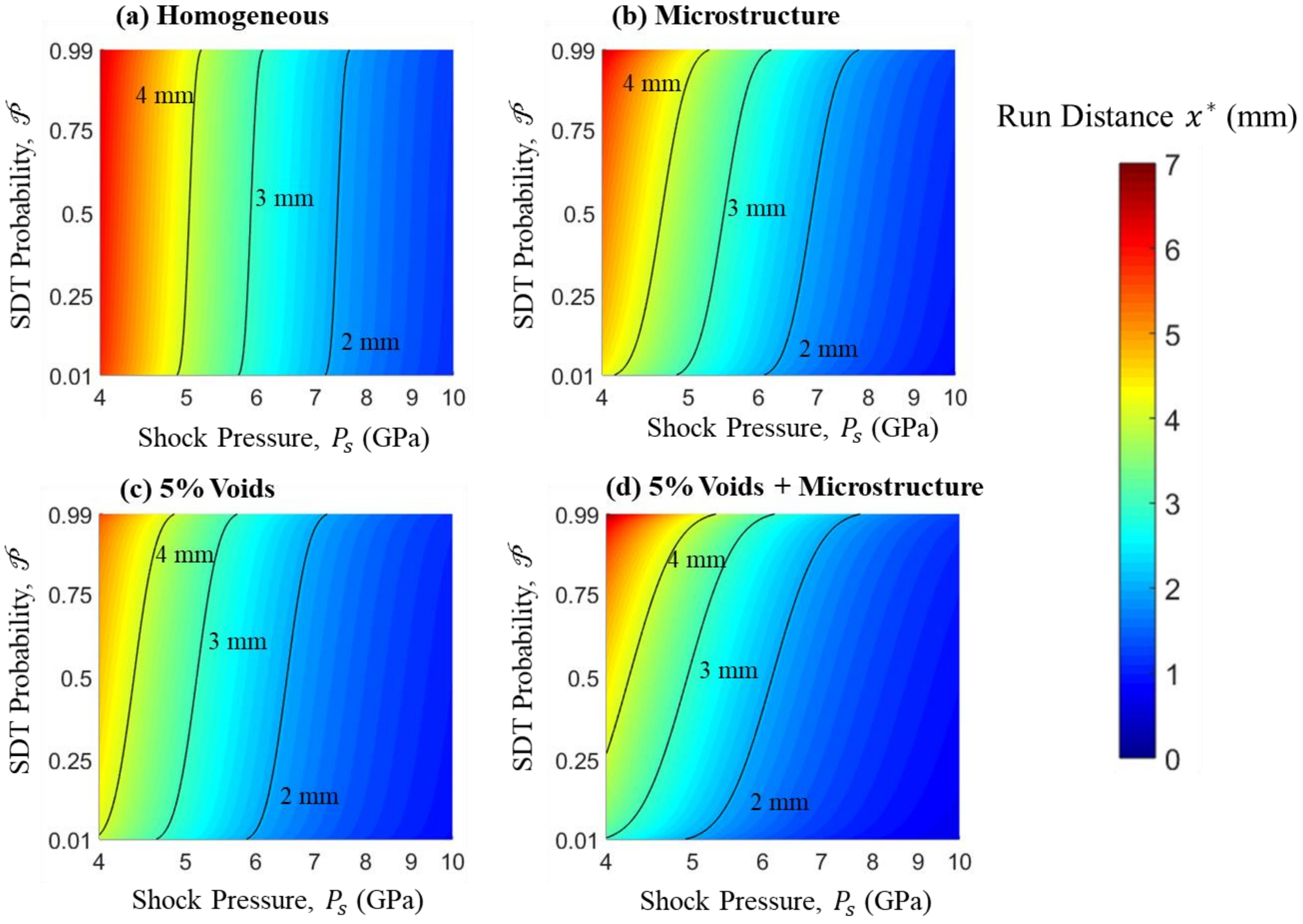
2D Pressure Map

$$P_s(\mathscr{P}, x^*) = \left\{ \frac{S}{\left(x^* - x_0^*\right)} \exp\left[\sqrt{2}\sigma_d \left(\mathrm{erf}^{-1}\left(2\mathscr{P} - 1\right)\right)\right]\right\}^{\frac{1}{m}} + P_0.$$

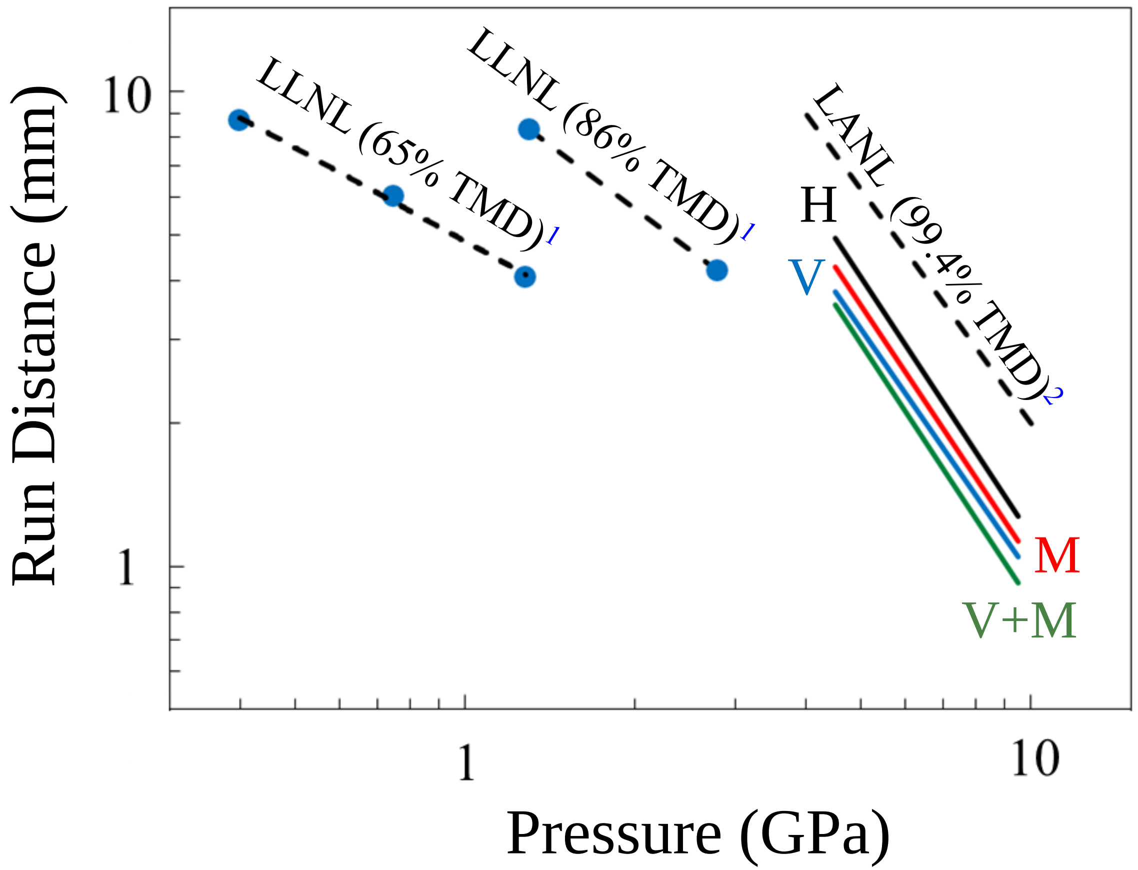


2D Run Distance Map

$$x^*(\mathscr{P}, P_s) = \frac{S}{\left(P_s - P_0\right)^m} \exp\left[\sqrt{2}\sigma_d \left(\mathrm{erf}^{-1}\left(2\mathscr{P} - 1\right)\right)\right] + x_0^*.$$



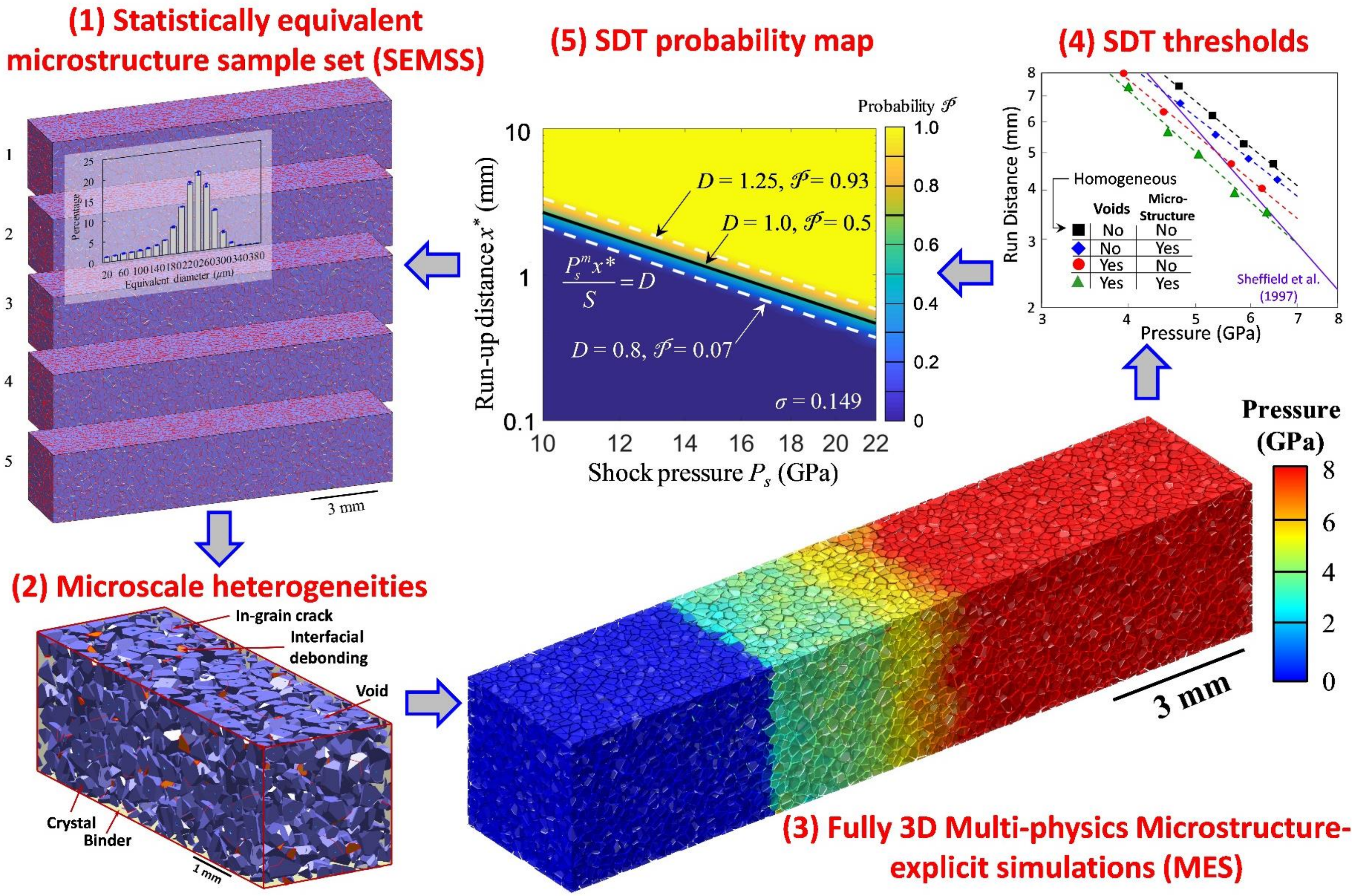
Comparison with Experiments



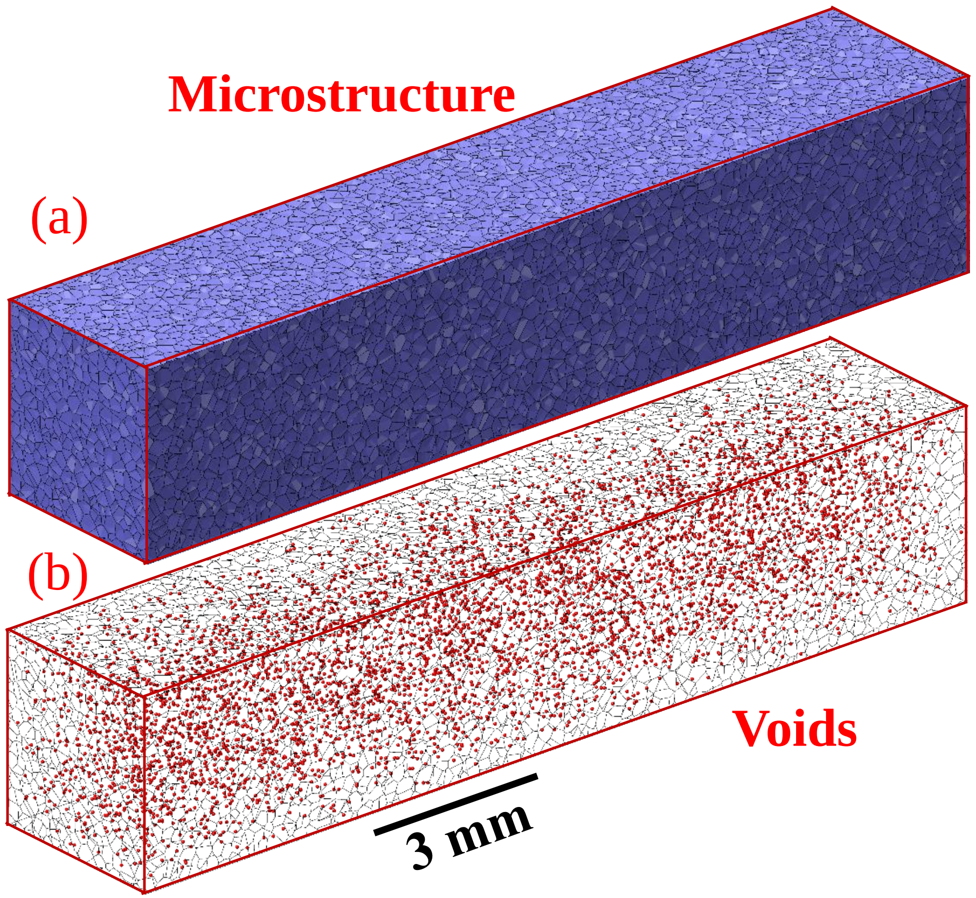
- Model trends match those seen in experiment.
- This model only represents a first approach at predicting SDT on a mm size scale.
- Further calibration of the HVRB or a transition to an Arrhenius-based chemical reaction rate model will be required.

1) Garcia, Frank, Kevin S. Vandersall, and Craig M. Tarver. "Shock initiation experiments with ignition and growth modeling on low density HMX." *Journal of Physics: Conference Series*. Vol. 500. No. 5. IOP Publishing, 2014.
2) Baytos, John F. *LASL explosive property data*. Vol. 4. Univ of California Press, 1980.

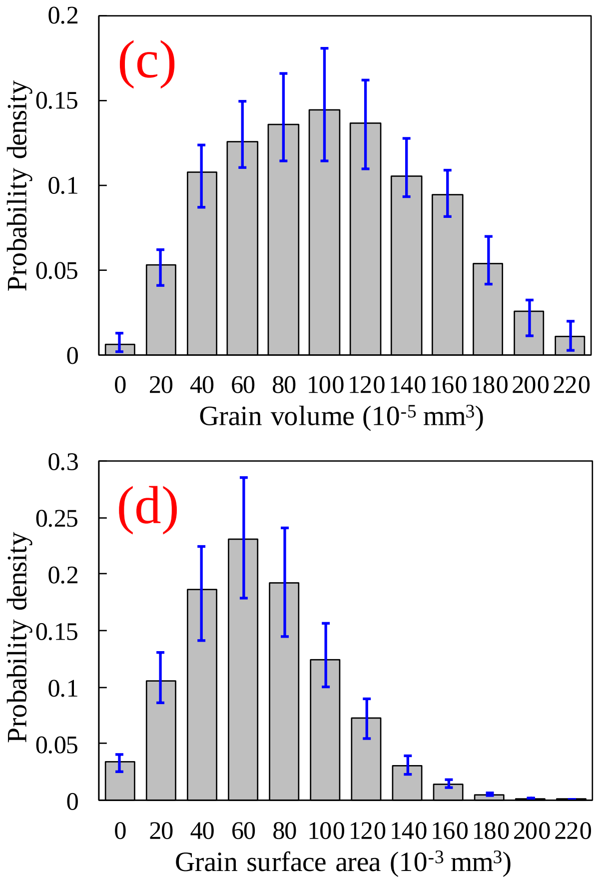
Design of Heterogeneous Energetic Materials (HEM) via MES



3D Microstructure-explicit/Voids-explicit Models (ME-VE-M)



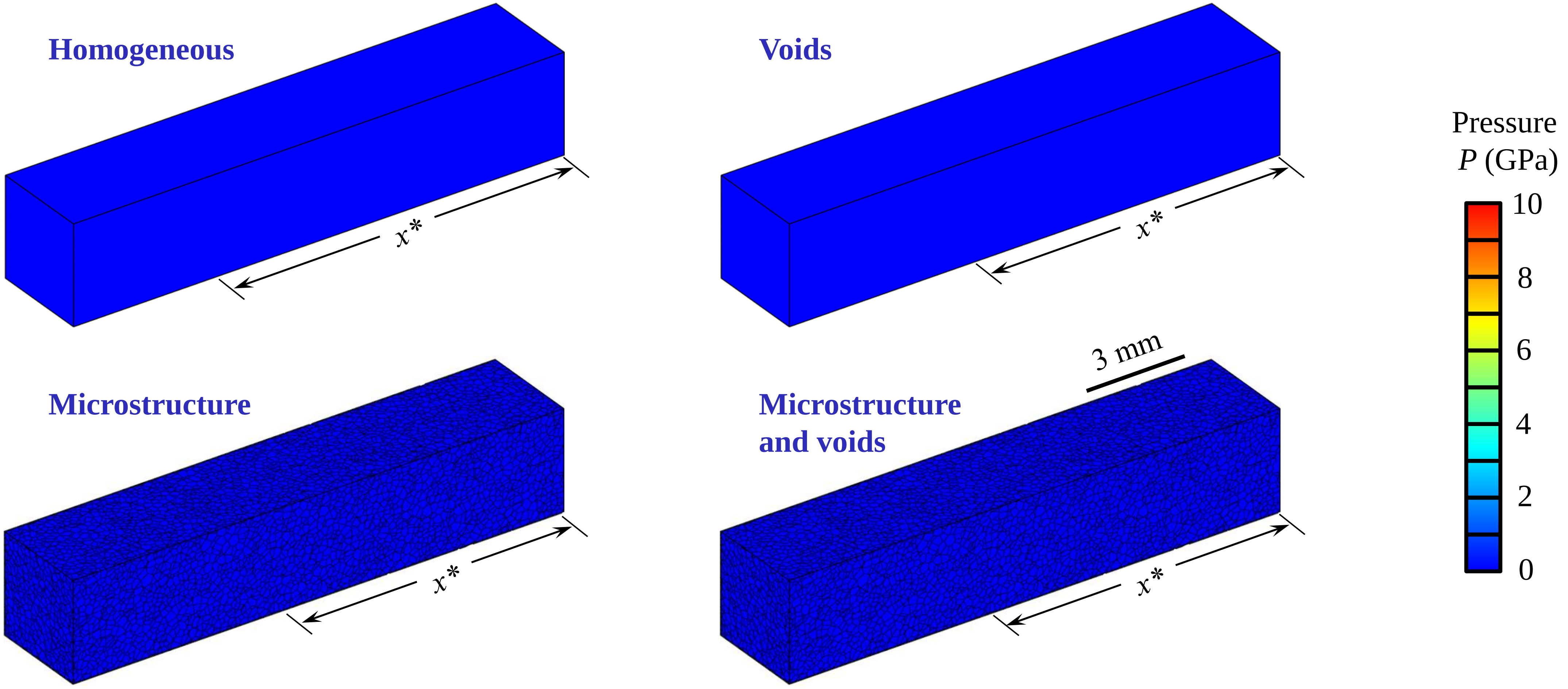
Grain size and surface area



- **Simulations performed using CTH – an Eularian-based hydrocode from SNL.**

ME-VE-Simulations of Shock to Detonation Transition (SDT)

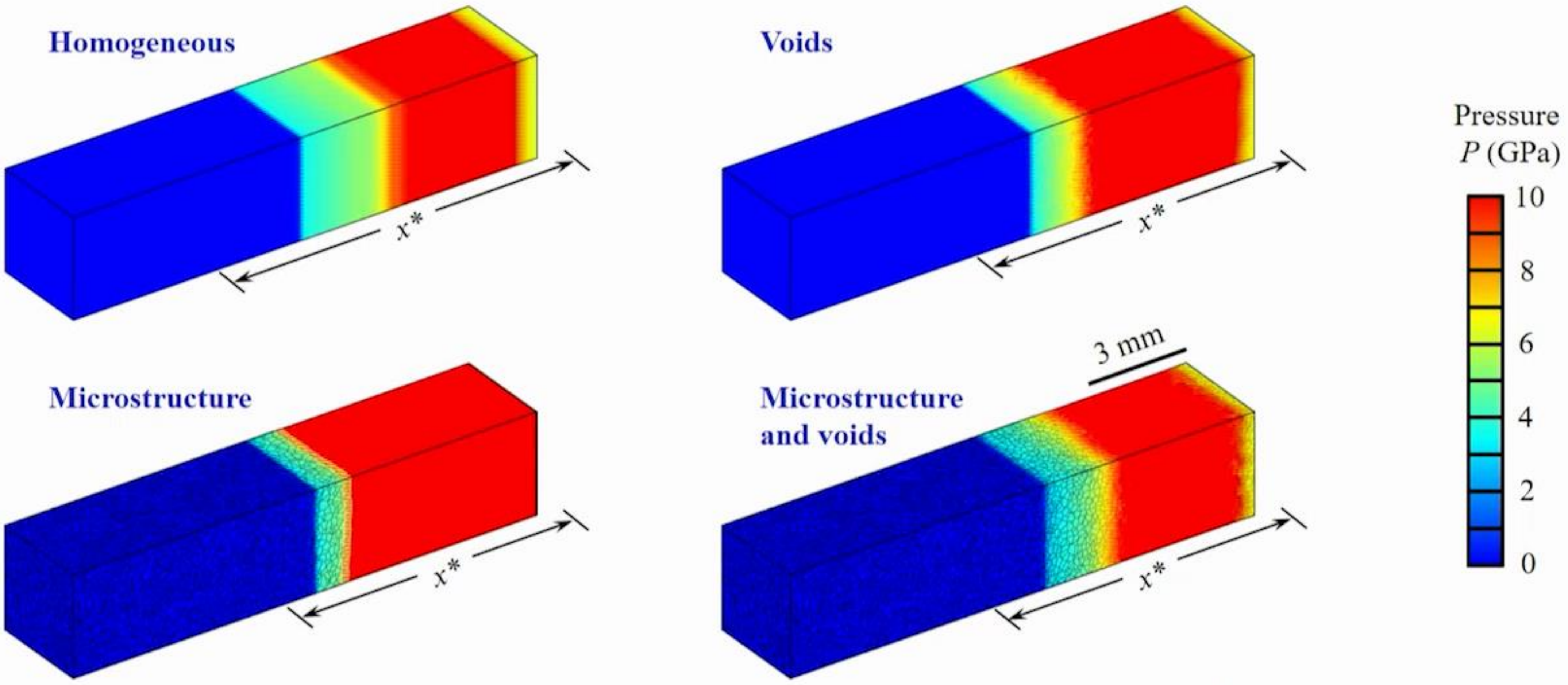
U_p = 600 m/s t = 0.00 μs



*x**: Run-to-detonation distance

ME-VE-Simulations of Shock to Detonation Transition (SDT)

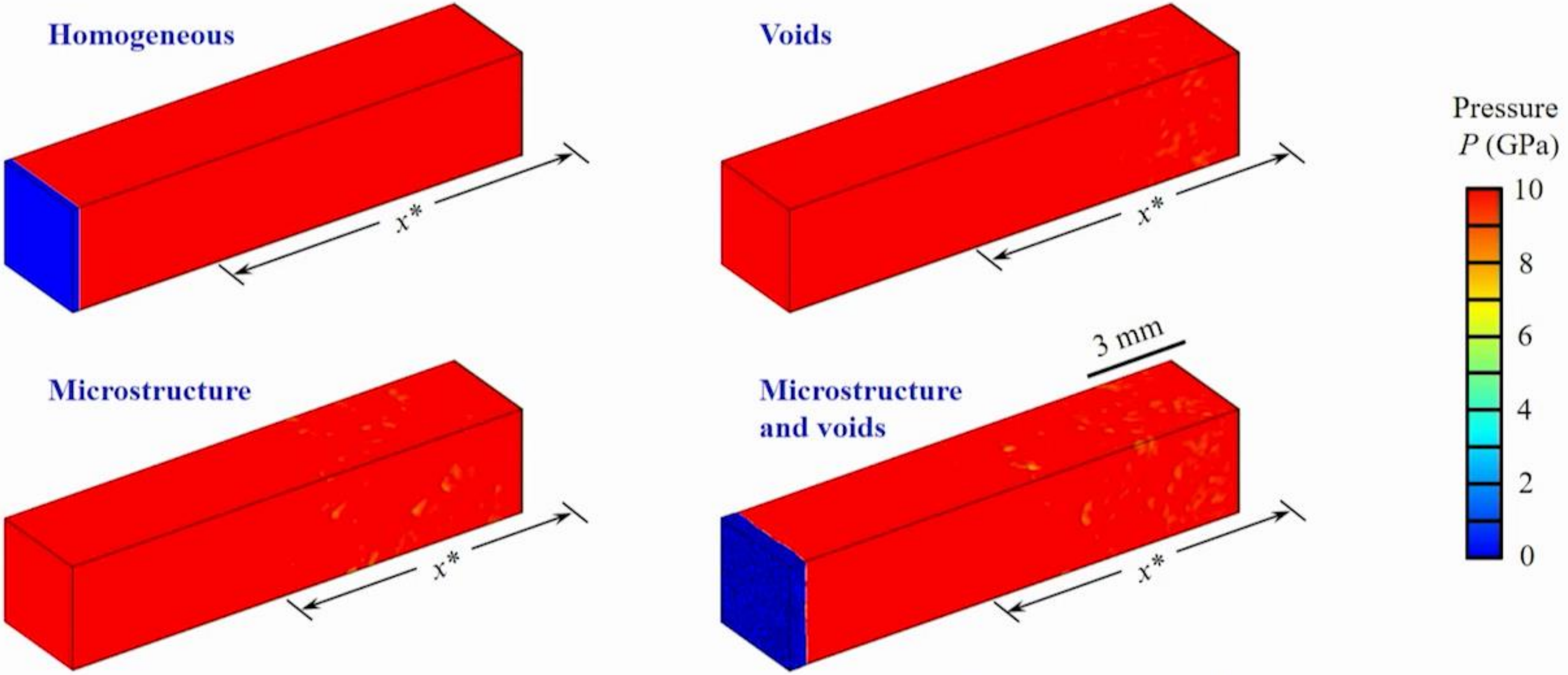
U_p = 600 m/s t = 2.08 μs



x^*: Run-to-detonation distance

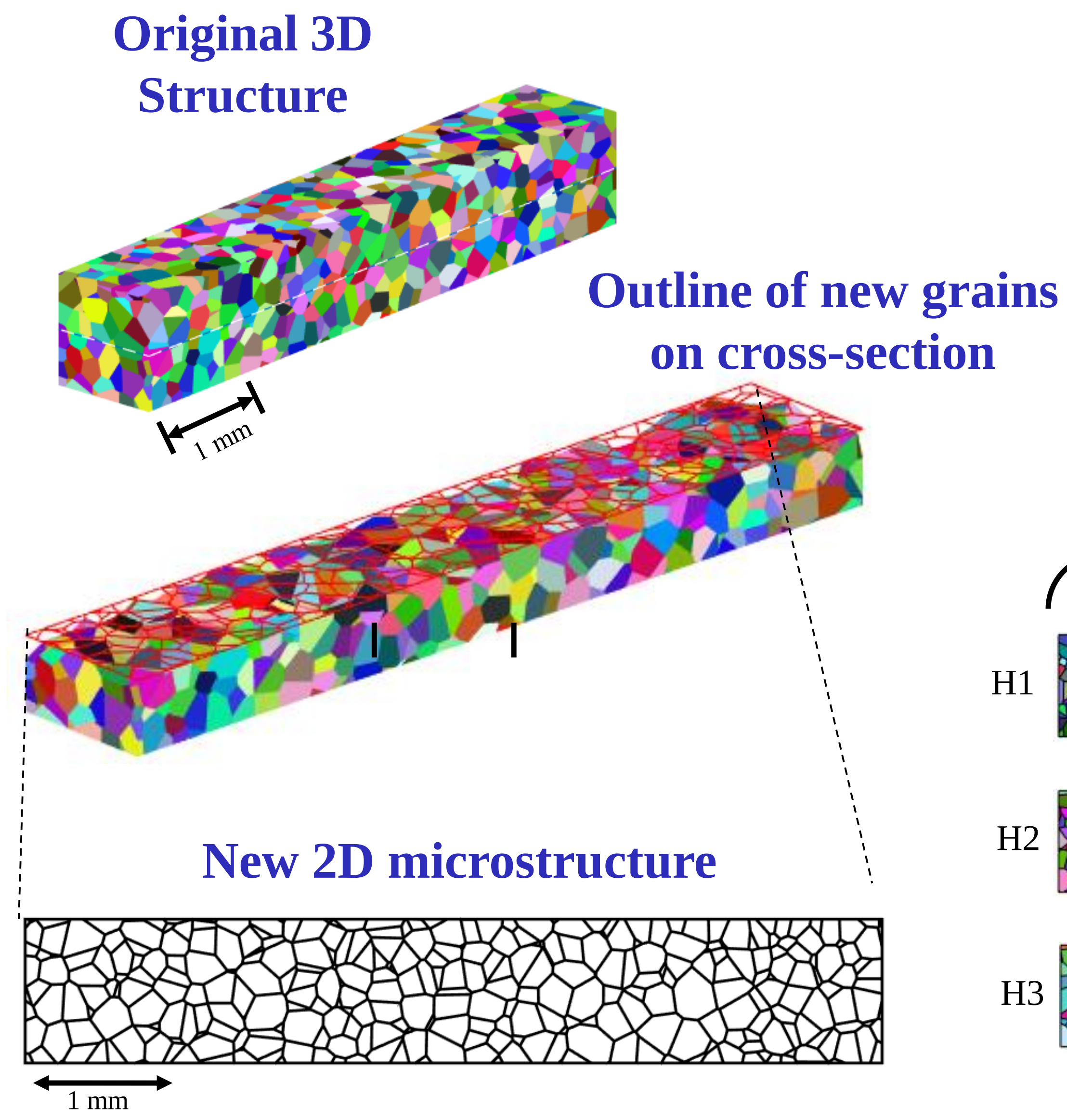
ME-VE-Simulations of Shock to Detonation Transition (SDT)

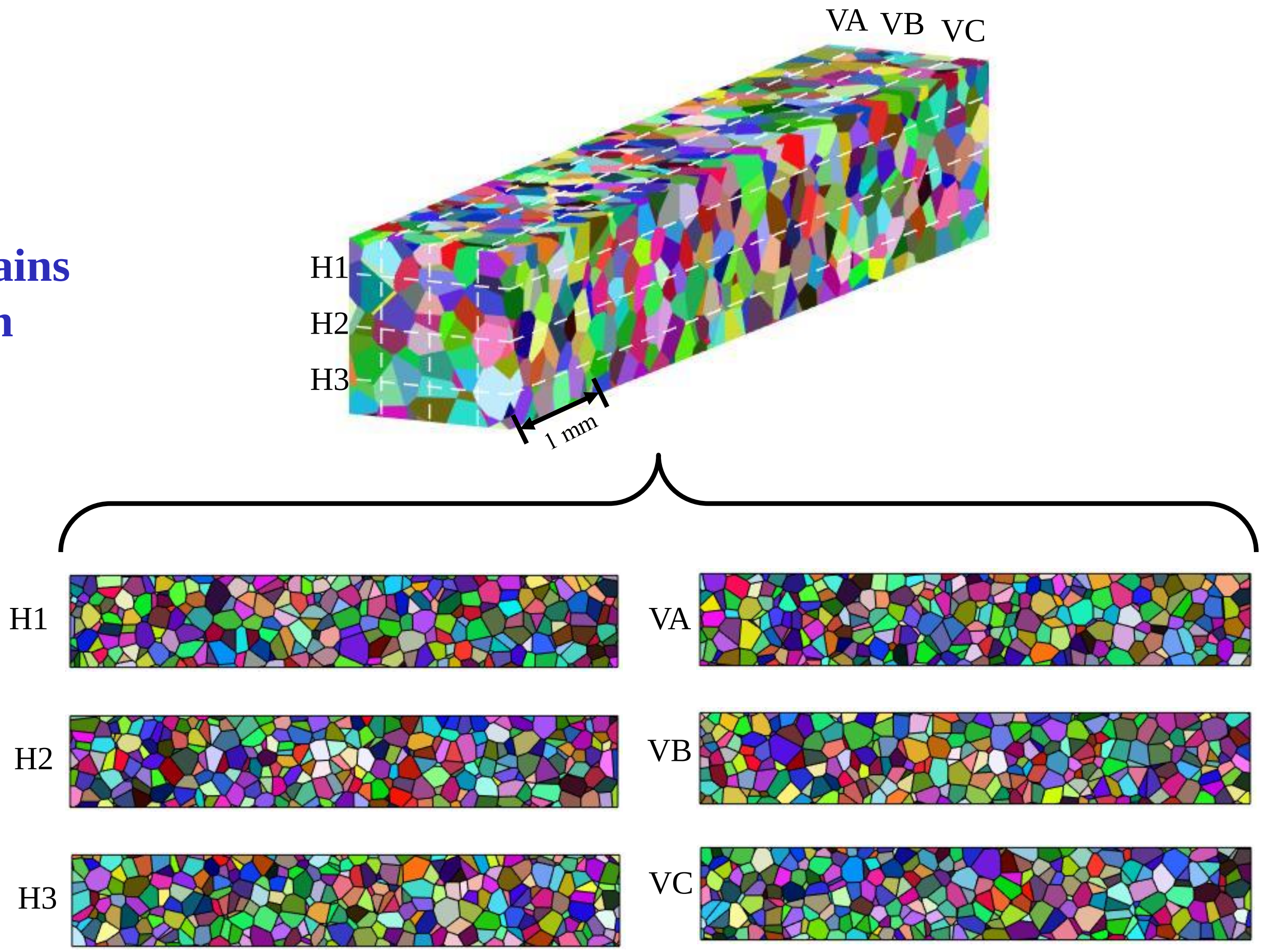
$U_p = 600$ m/s $t = 3.14$ μs



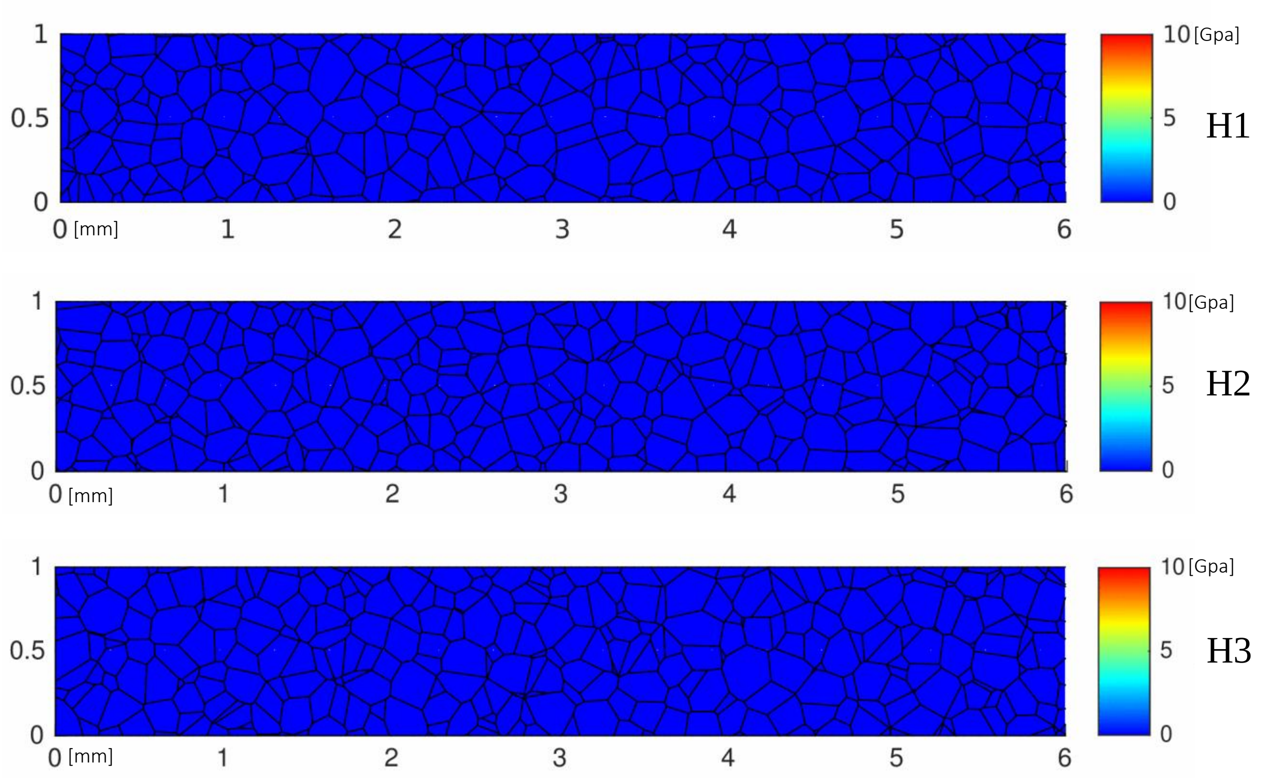
x*: Run-to-detonation distance

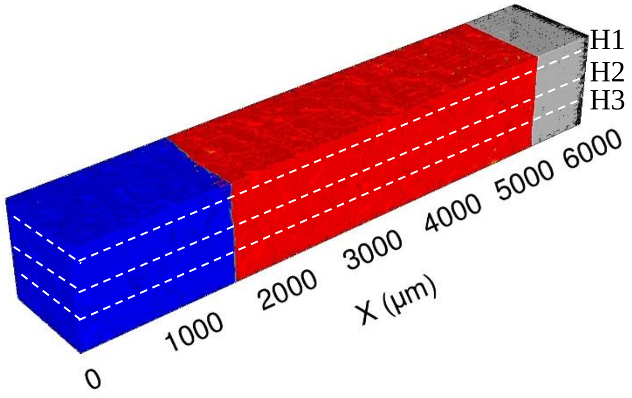
2D Sections of 3D Microstructure



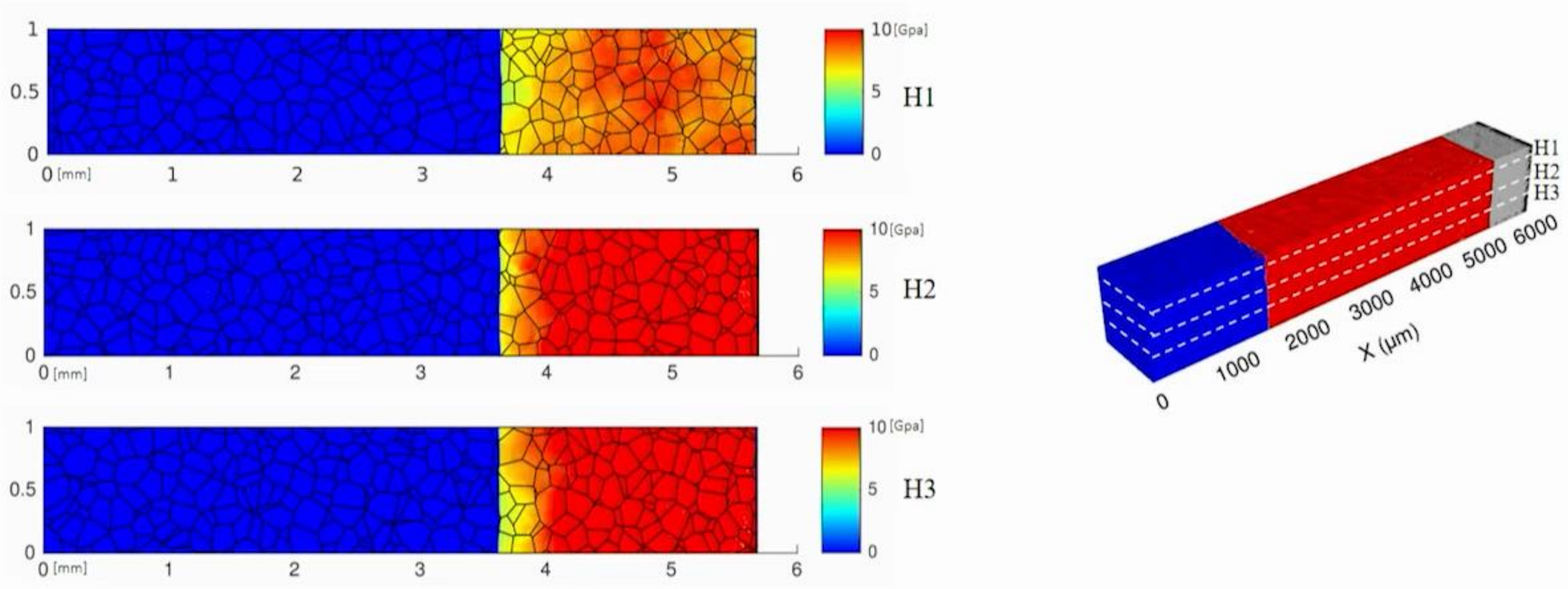


Pressure Fields: Horizontal (H) Sections

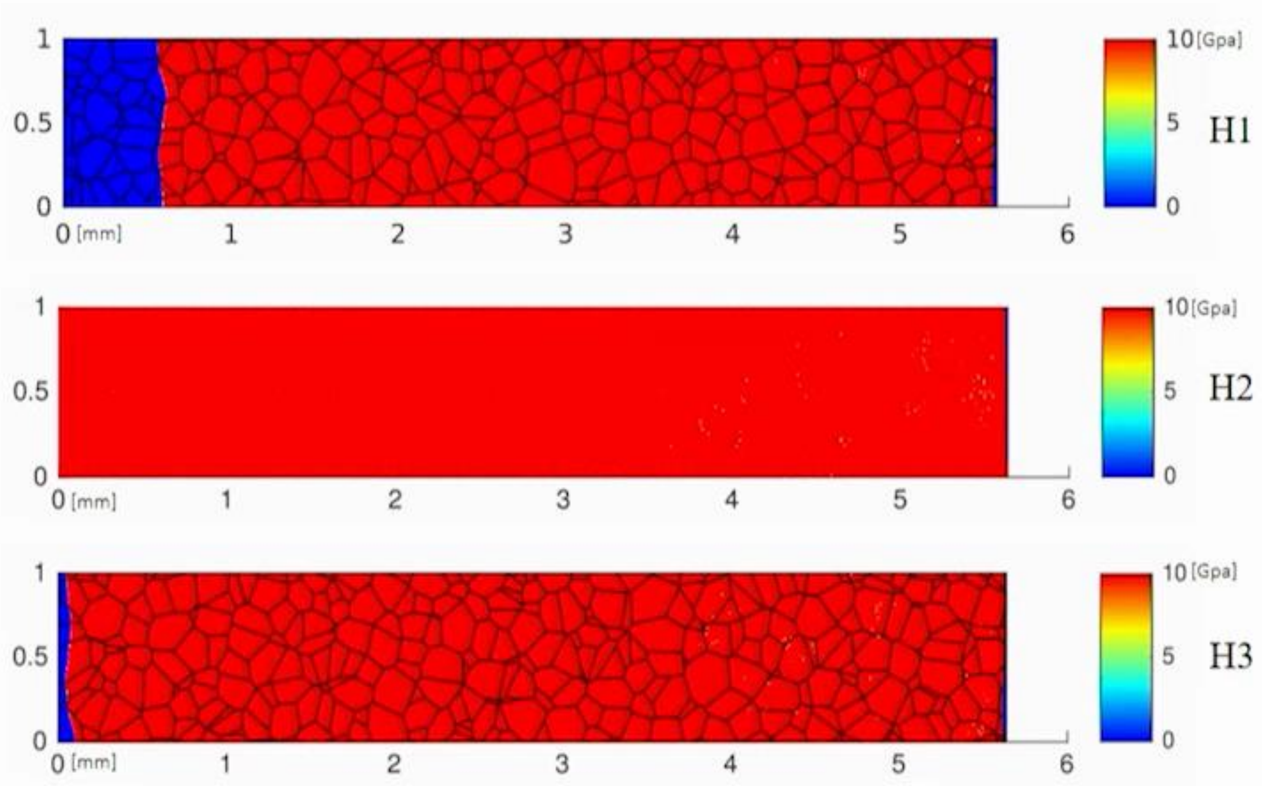


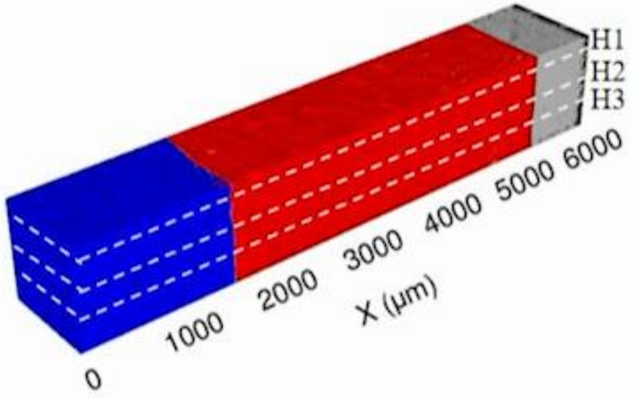


Pressure Fields: Horizontal (H) Sections

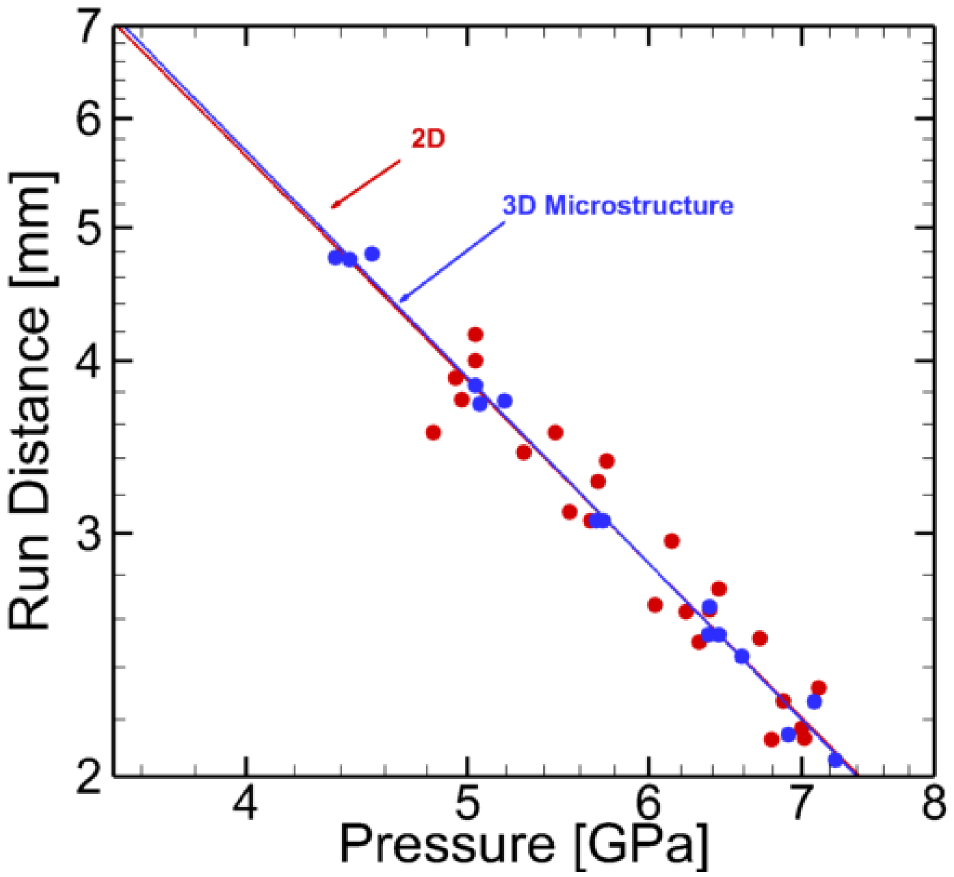


Pressure Fields: Horizontal (H) Sections



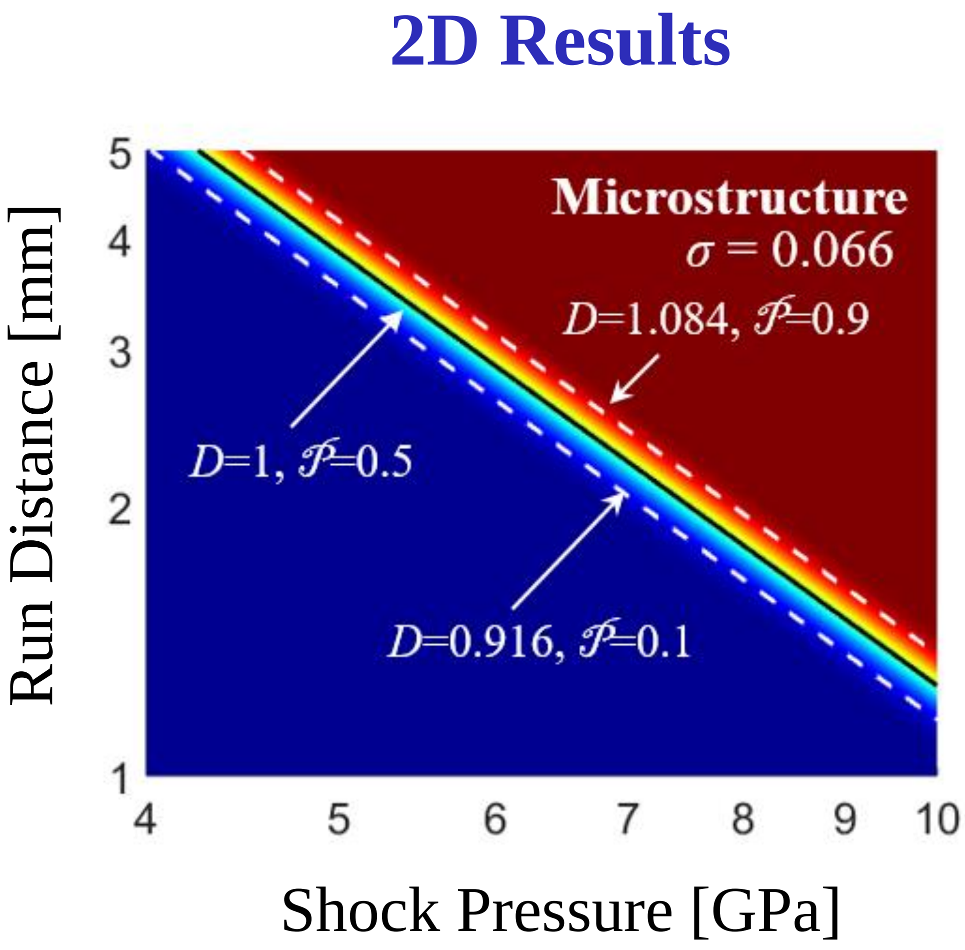


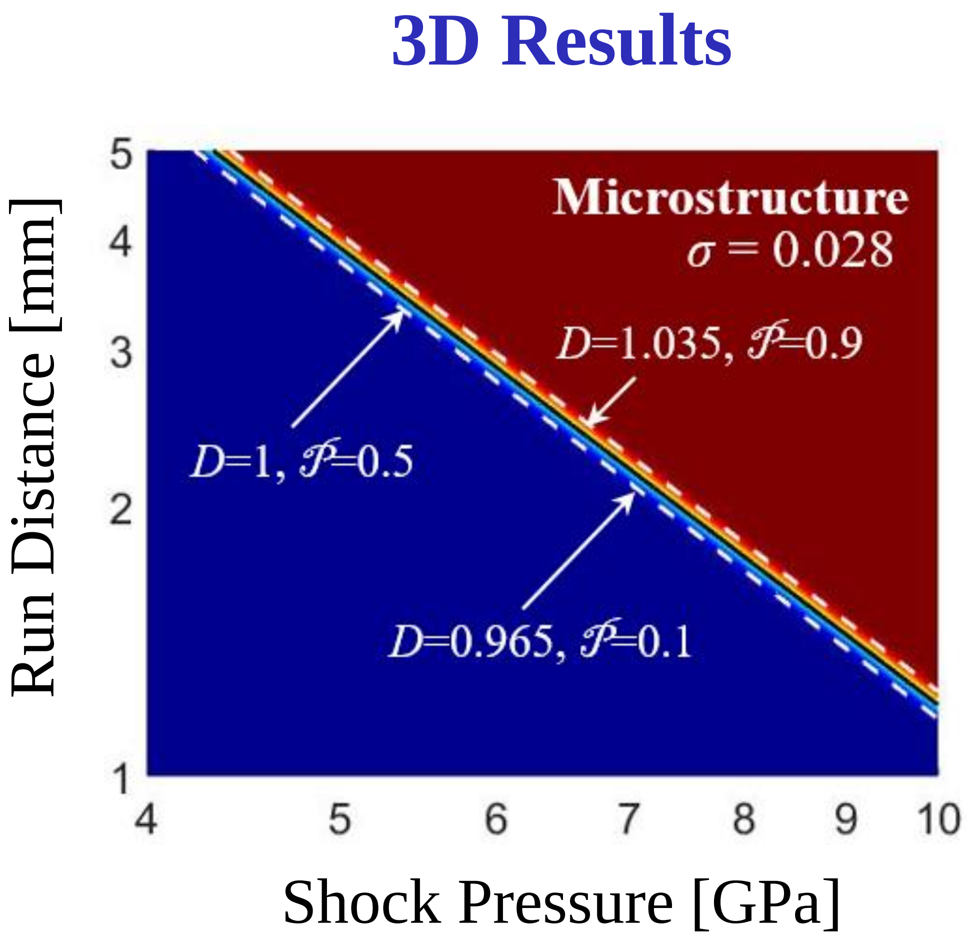
2D-3D Comparison

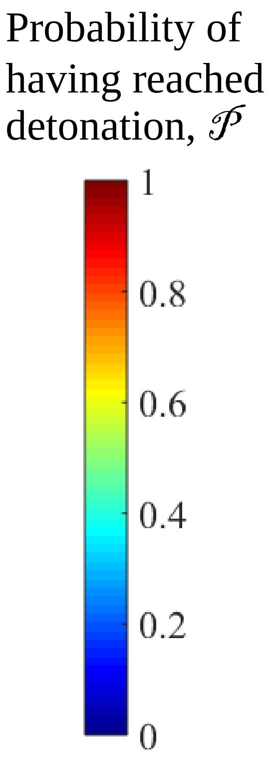


- Microstructure only case is chosen.
- There is overall agreement between the 2D and 3D results.
- 2D have more scatter than 3D, indicating that a single or smaller number of 2D runs may not accurately describe material behavior.

Probability Map Comparison





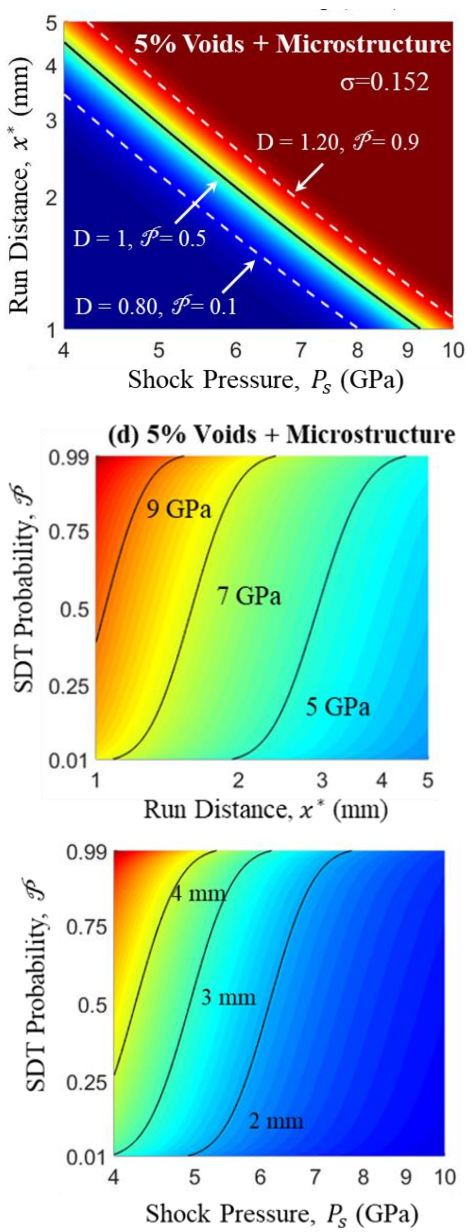


Summary

- MES (microstructure-explicit simulations) at mm size scales allow essential material effects to be captured for prediction of real material behavior.

- SEMSS allow prediction of probabilistic behavior assessment and UQ.

- Analytical relations for macroscopic behavior as functions of material structure for performance and sensitivity are being established and can be used for EM design.



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- All of my labmates for keep me sane these past five years!

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