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Shock compression of dense polymer and foam systems using molecular dynamics and DFT

J Matthew D Lane
Gary Grest, Aidan Thompson
Kyle Cochrane, Michael Desjarlais
Thomas Mattsson

Sandia National Laboratories
Albuquerque, NM

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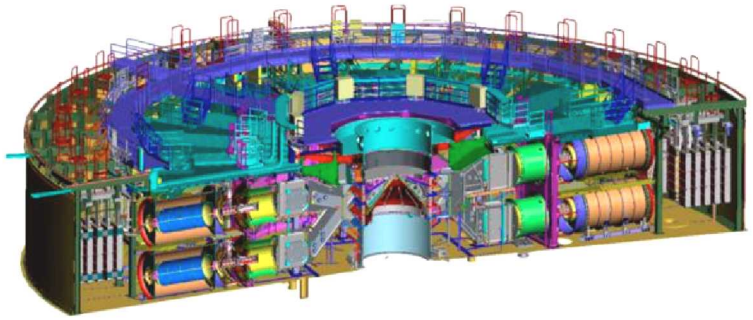
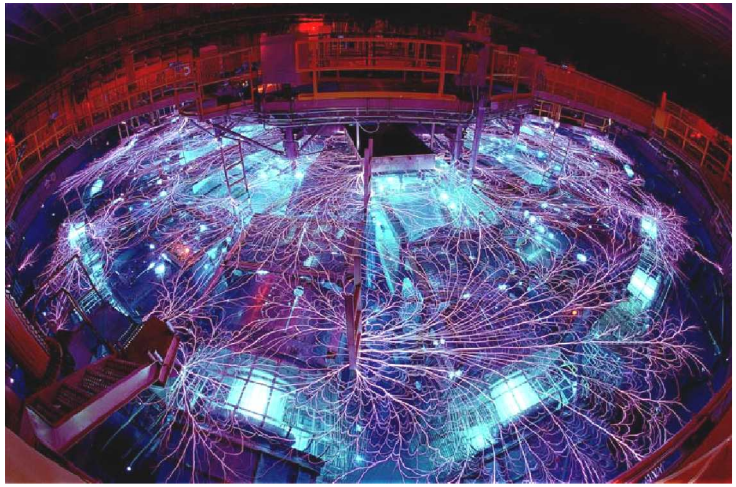
Motivation for polymer shock simulation

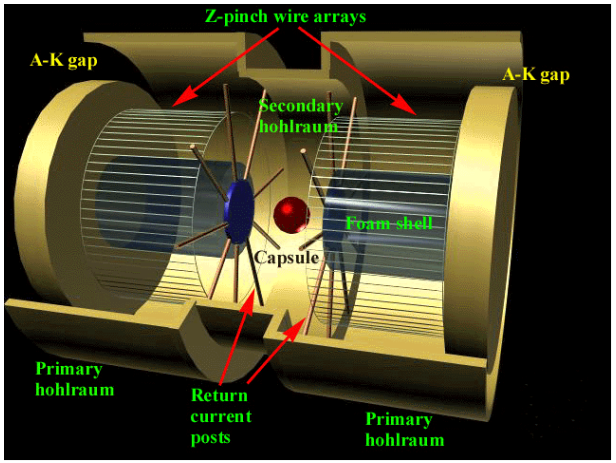
Z-machine at Sandia National Labs





Z-machine experiments allow:

- Extremely high-pressure shock studies
- Inertially confined fusion (ICF) research
- Better understand EOS for polymers and mixtures



Double-ended Z pinch with a cm sized foam-shell capsule

Matthew Lane - jlane@sandia.gov

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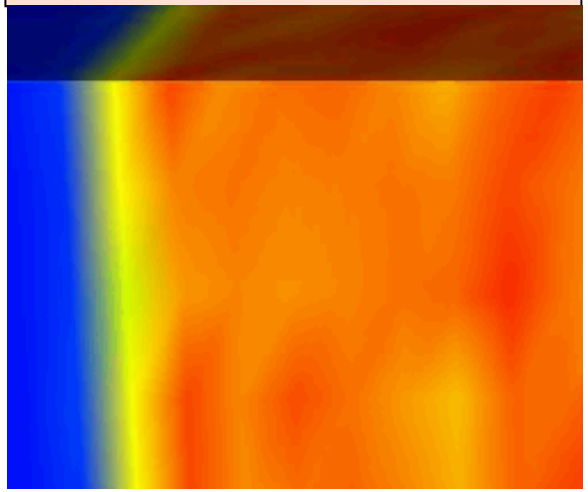
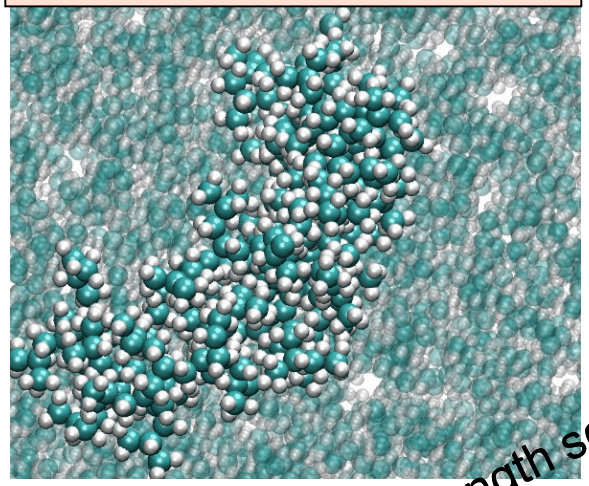
Hierarchy of modeling approaches

Foams response depends on processes/structure at several lengthscales:

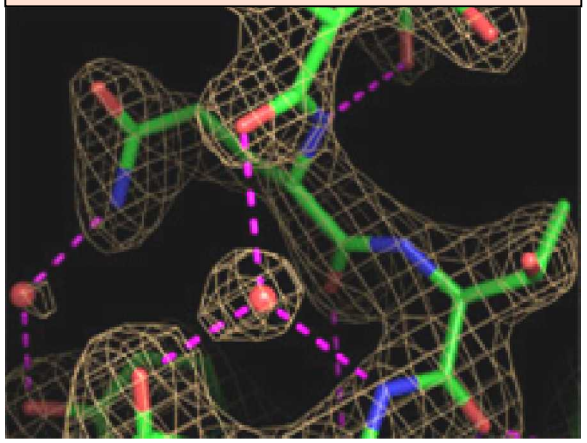
Continuum scale
μs times & nm-mm lengths



Molecular scale
ns times & 10s nm lengths

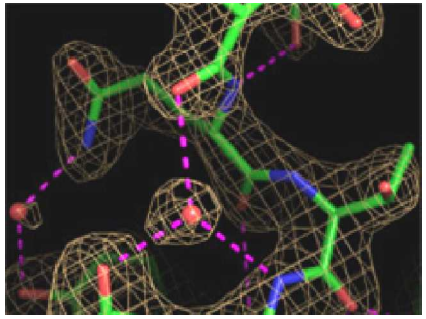


Quantum scale
fs times & Å lengths

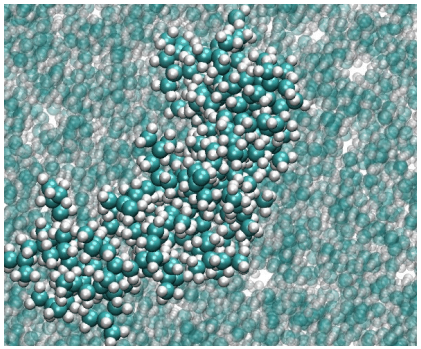


Increasing time and length scales

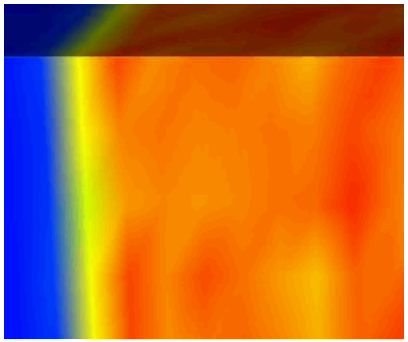
Hierarchy of modeling approaches



- VASP 5.1 code (Georg Kresse, Vienna, Austria)
- Plane-wave basis-set allows controlled convergence & free electrons/ionization
- Finite-temperature DFT with Projector augmented wave core functions (PAW)
- Sandia developed AM05 functional (Armiento & Mattsson, Phys Rev B 2005)
- Kyle Cochrane, Session M5.00004 on Wednesday afternoon

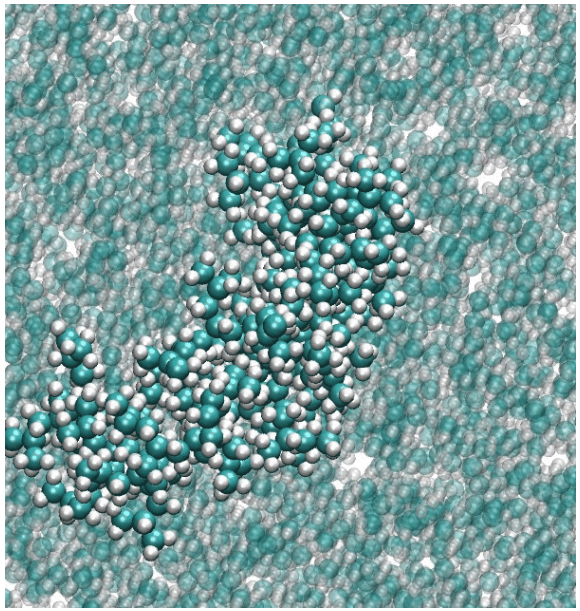


- LAMMPS code (Steve Plimpton, et al., Sandia National Labs)
- MD engine for atoms, molecules, or particles at any length/time scale
- Multiple common interatomic potentials allows for comparison
- Integrated MPI for massively parallel simulations
- http://lammps.sandia.gov
- This talk



- ALEGRA code (Sandia National Labs)
- MHD engine for 3D modeling of continuum systems
- Mesoscale response with multiple physics models
- http://www.cs.sandia.gov/ALEGRA
- Thomas Haill, Session L3.00004 on Wednesday morning

Dense polymer simulation



T.R. Mattsson, et al., *Phys Rev B*, 81, 054103 (2010).

- Two polymers:

Polyethylene - simplest possible linear carbon backbone structure forms semi-crystalline solids

PMP (TPX) - branched hydrocarbon with bulky side chains which is good for producing amorphous foams

Molecular Dynamics

OPLS, Jorgensen et al., JACS, 118, 11225 (1996)

Borodin-Smith exp-6, JPCB, 110, 6279 (2006)

AIREBO, Stuart et al., JCP, 112, 6472 (2000)

ReaxFF, vanDuin et al., JPCA, 112, 1040 (2008)

- 22,000+ atom PE sample and 45,000+ atom PMP
- Uniaxial Hugoniotstat method employed to compress

Density Functional Theory

AM05 Functional

- Several hundred atom samples of PE and PMP

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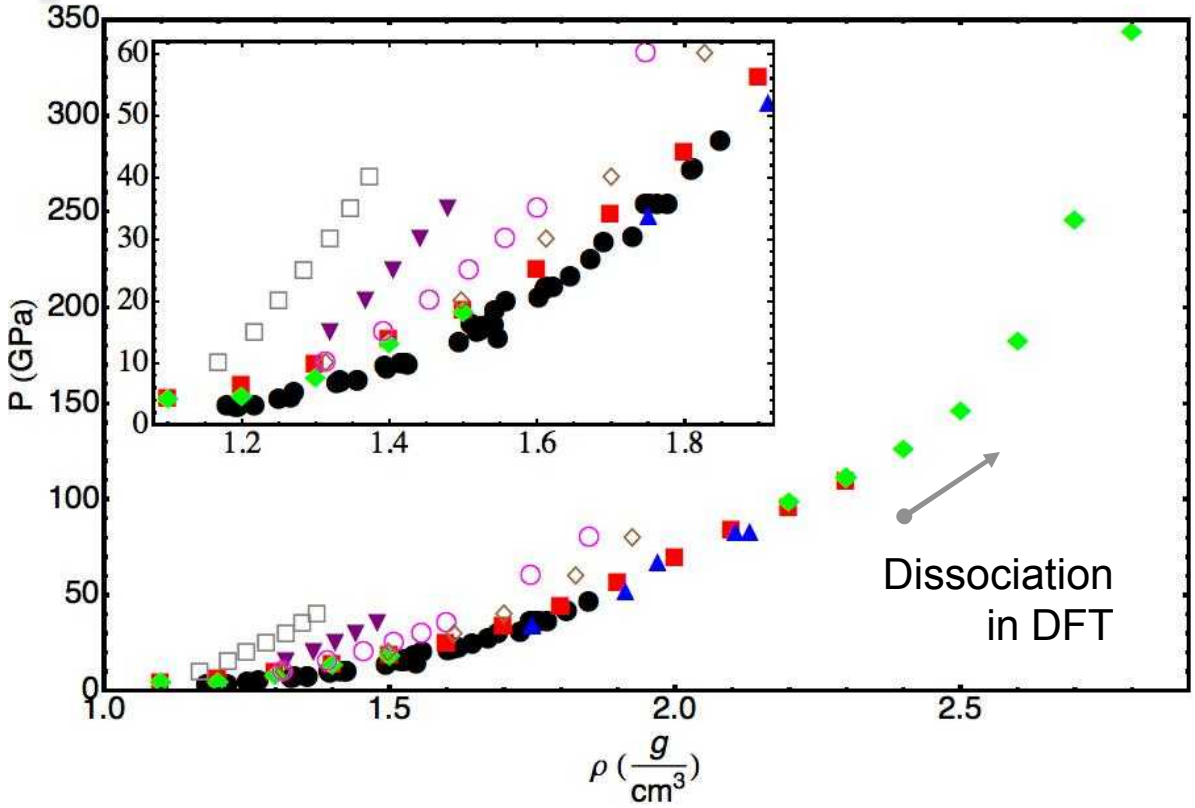
Polyethylene shock Hugoniot



- AIREBO
- OPLS
- Borodin-Smith (exp6)
- ReaxFF
- DFT-AM05
- Experiment (Nellis/LASL)

T.R. Mattsson, et al., *Phys Rev B*, 81, 054103 (2010).

- AIREBO and OPLS both give significantly too stiff shock response at all pressures
- Borodin-Smith and ReaxFF better choices for weak shocks in polyethylene
- Only the DFT-AM05 simulation of high fidelity also for strong shocks
- Significant deviations already well before the regime where dissociation occurs
- Reactive properties of force-fields are not important for weak shocks

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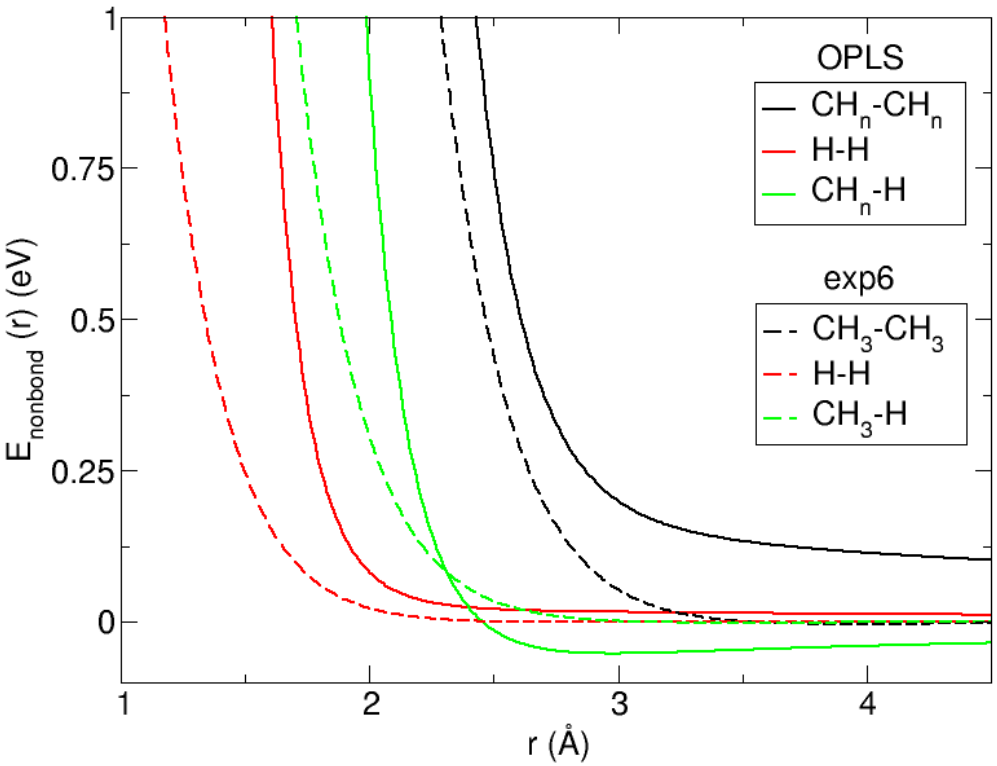
Core repulsion model is critical



- AIREBO and OPLS share L-J 12-6 functional form for repulsive interaction
- Borodin-Smith exp-6 has a weaker exponential form, more physical shock behavior
- Shock-problems probe regions of the potential far from the equilibrium region -- where they are parameterized and exhibit very similar characteristics
- *Behavior under shocks are difficult to predict from equilibrium properties*

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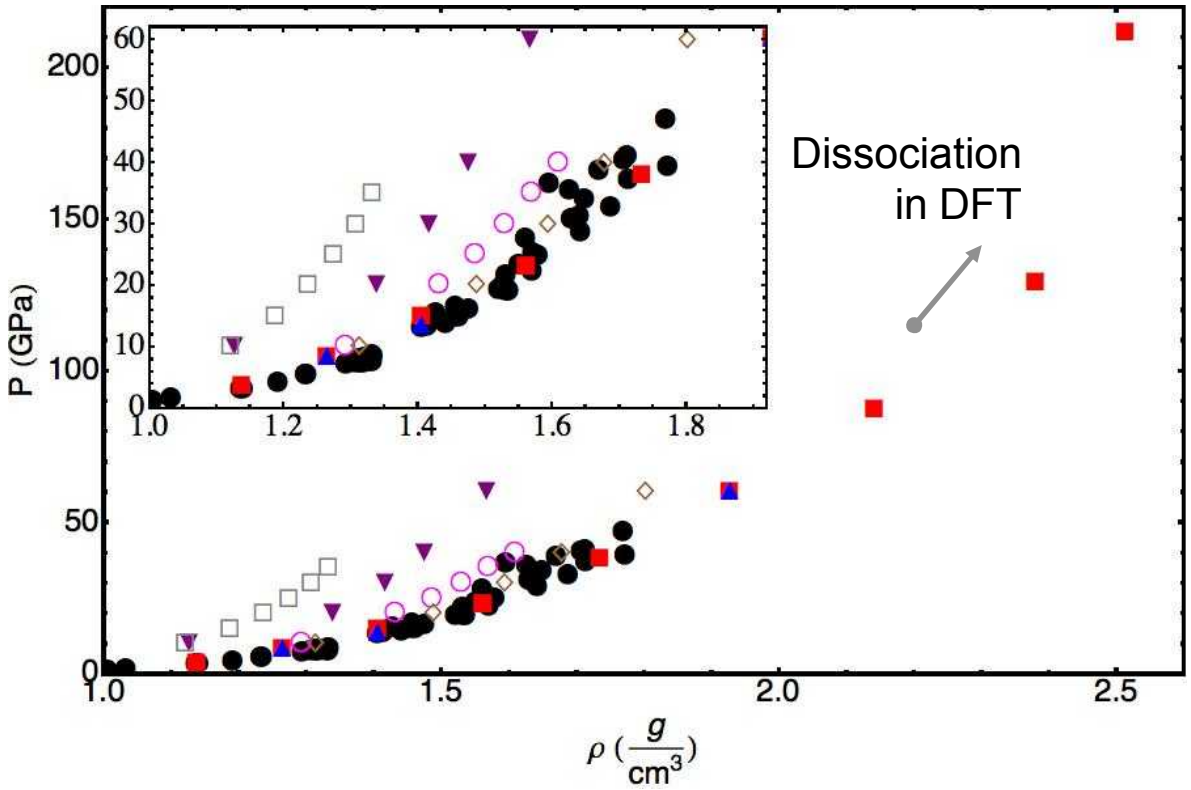
Poly(4-methyl 1-pentene) shock Hugoniot



- AIREBO
- OPLS
- Borodin-Smith (exp6)
- ReaxFF
- DFT-AM05 (struct A/B)
- Experiment (LASL)

T.R. Mattsson, et al., *Phys Rev B*, 81, 054103 (2010).

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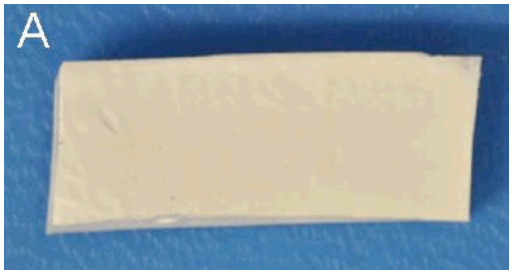
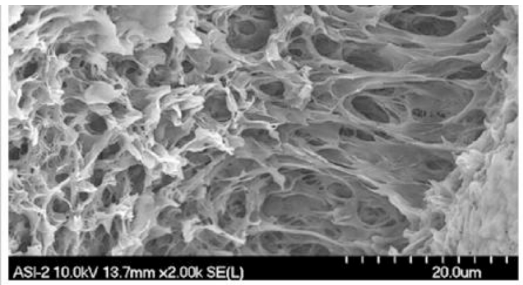
Foam introduction





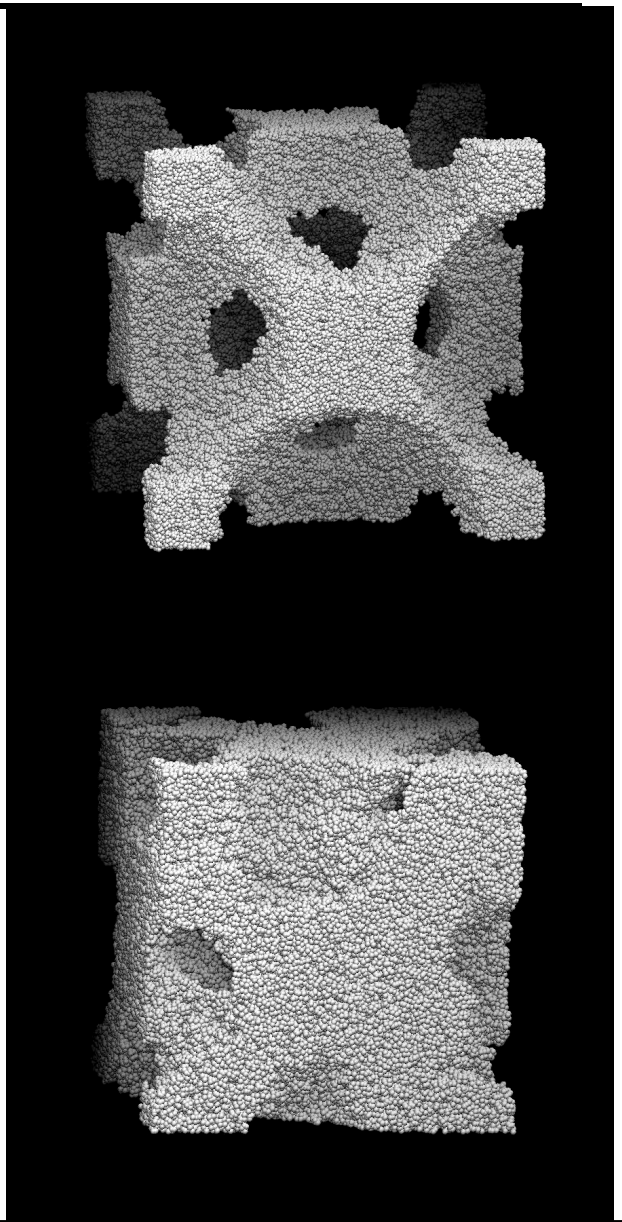
Densities
- Experimental foam – 0.309 +/- 0.025 g/cc
- Simulated foam – 0.300 g/cc

Experimental fabrication:
- 0.833 g/cc PMP dissolved in cyclohexane & evaporated

Simulation fabrication:
- 0.833 g/cc PMP perforated with growing spherical indenters placed on an fcc lattice, creating a unit cell of foam with unit cell of 20x20x20 nm.

ReaxFF potential was used exclusively.

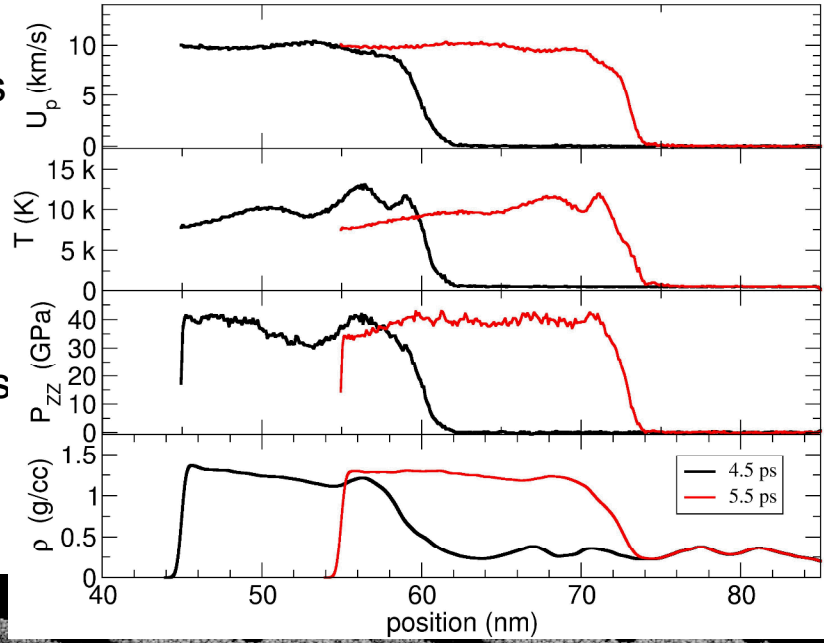
S. Root, et al., (to be submitted 2011).

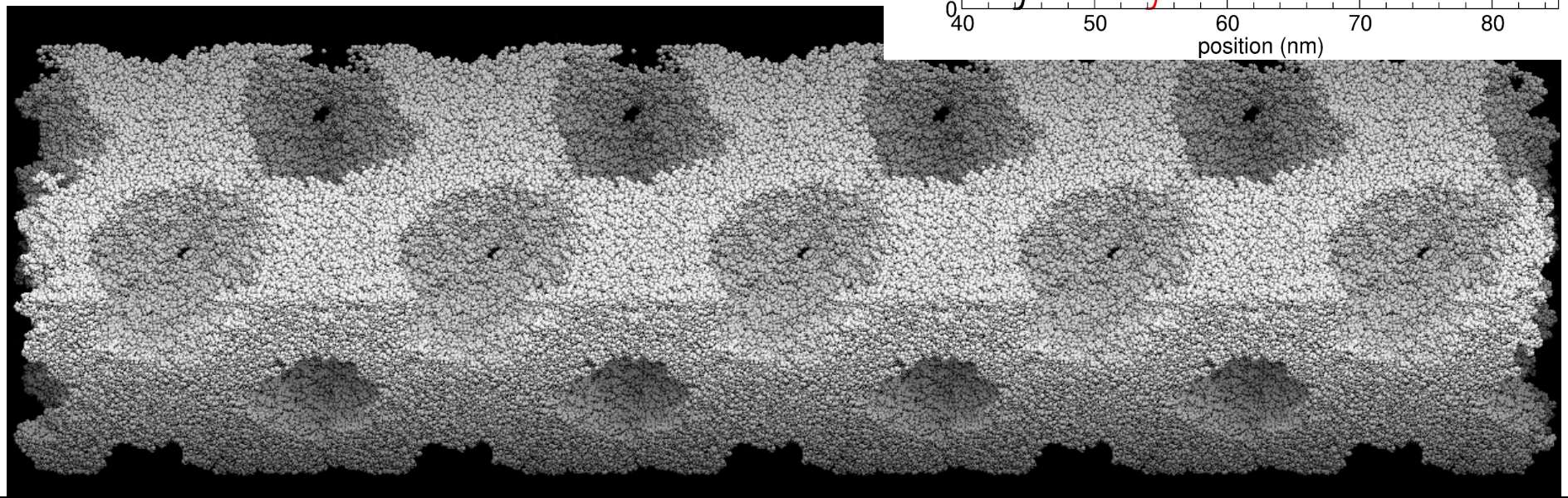


Matthew Lane - jlane@sandia.gov

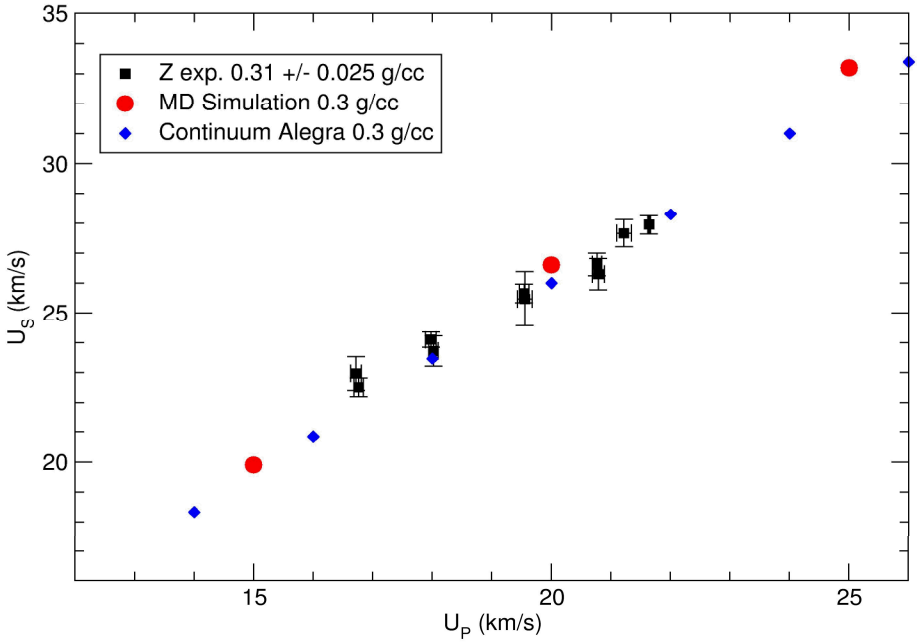
Foam shock methodology

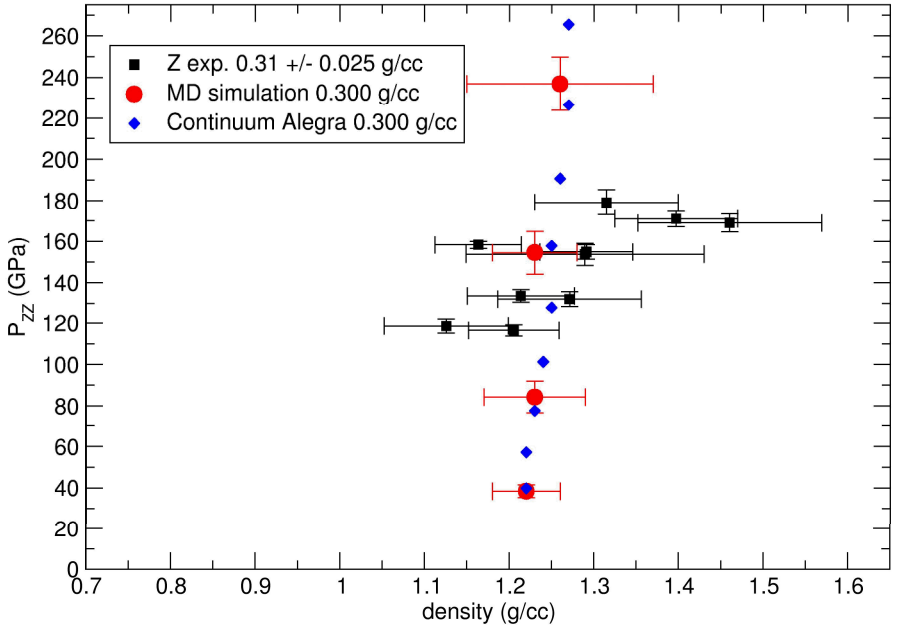
- Shock Hugoniotstat method could not be used, so long simples were constructed from foam unit cells
- Shock is driven by warm momentum mirror using standard NEMD techniques.
  - Piston velocities ranged from 10 to 30 km/s.
  - Very short simulation timesteps (0.01 to 0.05 fs were required
- Profiles were calculated from per-atom variables





Foam results – Hugoniot data





- Good quantitative agreement between Z experimental data, and both MD and continuum simulations for ~ 0.310 g/cc foam shock loading

- Spread in experimental data comes from variation is sample density and inhomogeneity.

Matthew Lane - jlane@sandia.gov

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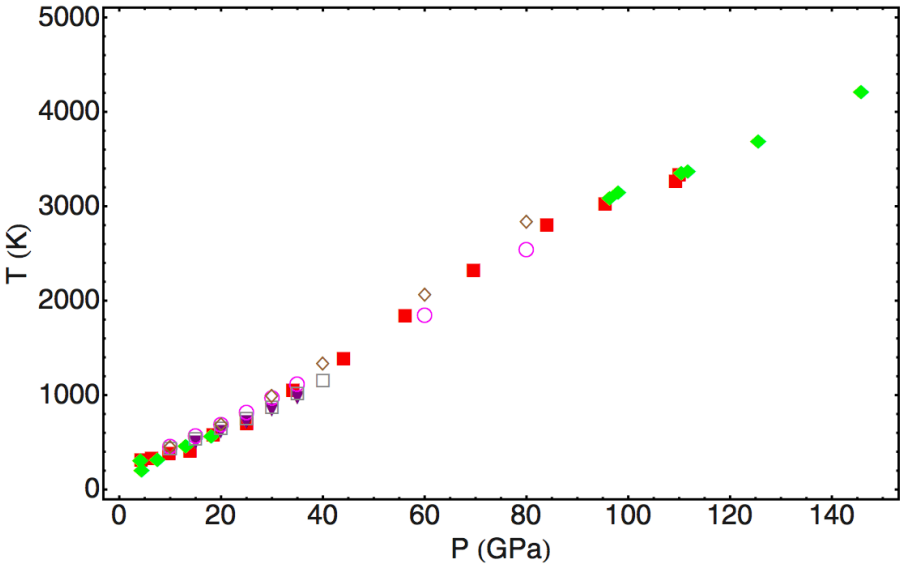
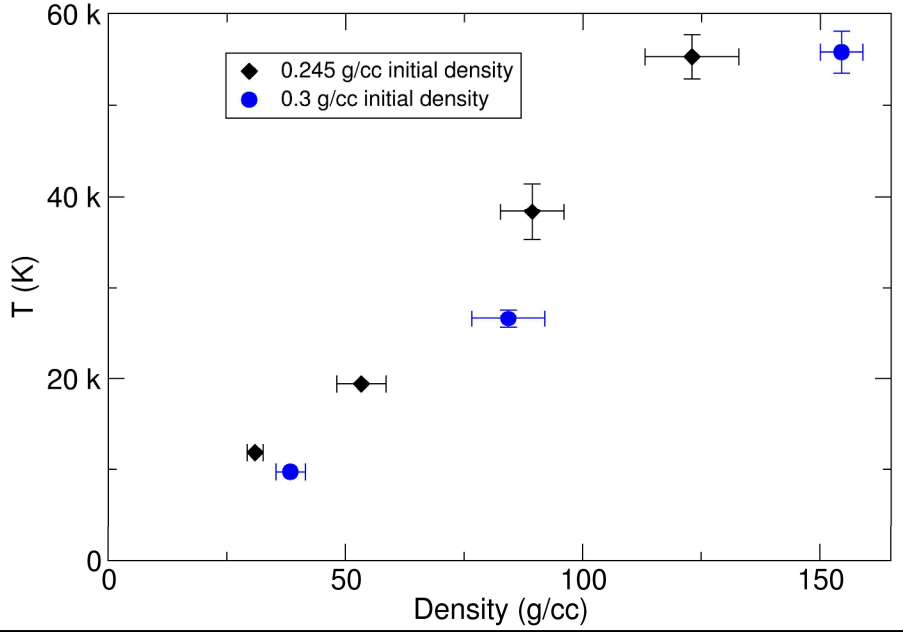
Temperature dense polymer vs foam



Temperatures in dense polymer were < 5000 K through 150 GPA

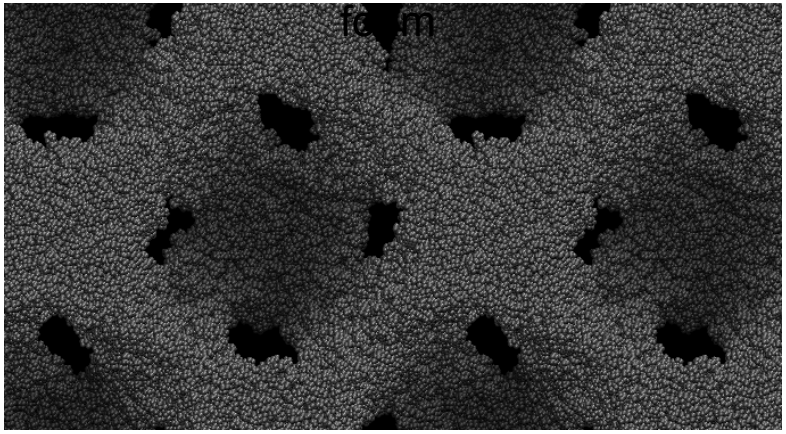
Temperatures in foams spike comparatively higher, driving dissociation at much lower densities and pressures



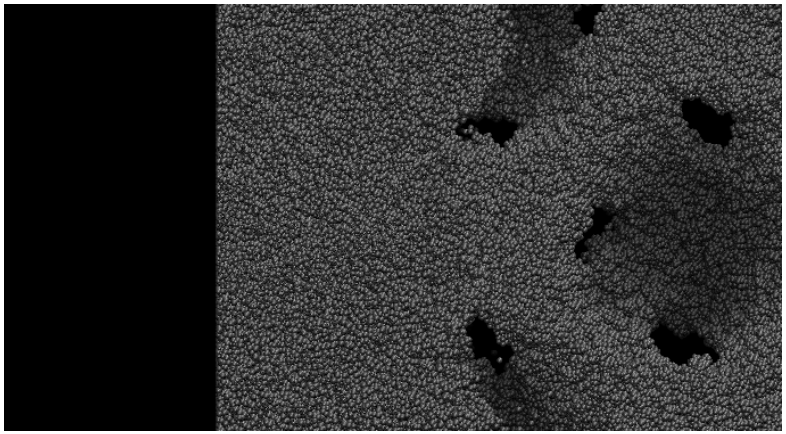
Foam results – dissociation and vapor

10 km/s piston – 0.300 g/cc PMP

Initial state

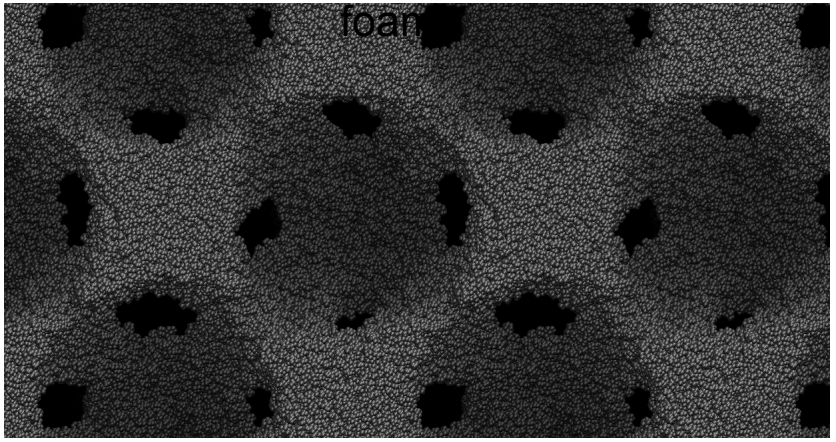


Shocked state

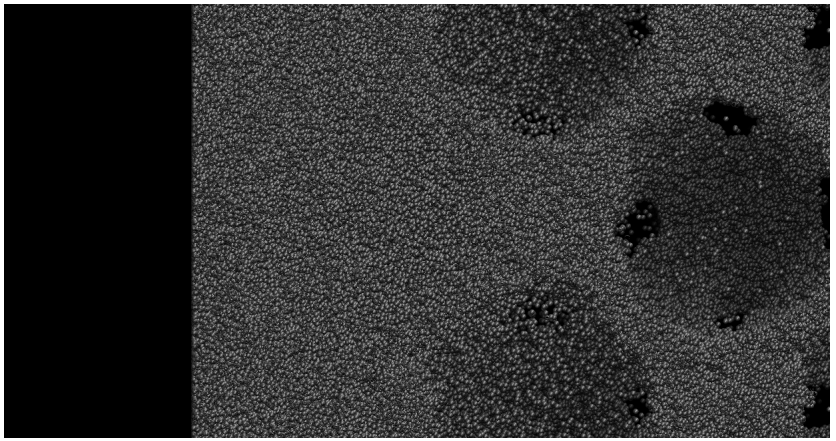


30 km/s piston – 0.300 g/cc PMP

Initial state



Shocked state



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Summary

- For dense polymers, shock response depends more on accurate potentials than accurate structure. For this reason, DFT is an excellent tool in dense polymer studies

- MD potentials exist, such as ReaxFF, which capture the quantitative shock response of both dense polymers and foams up to 50 GPa in dense polymers and higher in foams

- Good agreement was found between experiment and MD simulations of polymer foam. But, there is still much to resolve in determining the role of inhomogeneity, detailed structure and length scales on shock response.