US-Russia 6-Lab meeting, Barcelona, Spain, Octob[illegible]

SAND2011-7821C

# Shock compression of Hydrocarbon Polymers:

## A molecular dynamics approach with comparison to DFT, Continuum and Experiment


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Albuquerque, New Mexico



SAND 2011-



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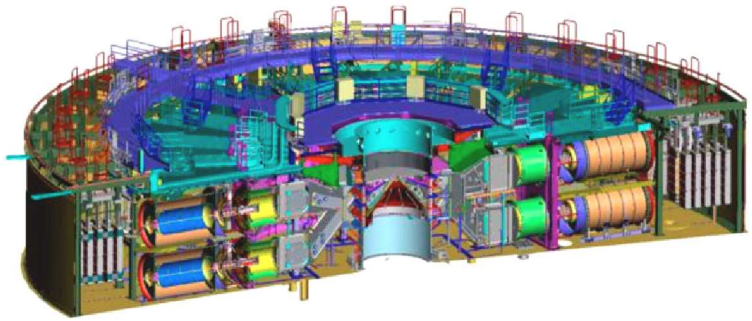
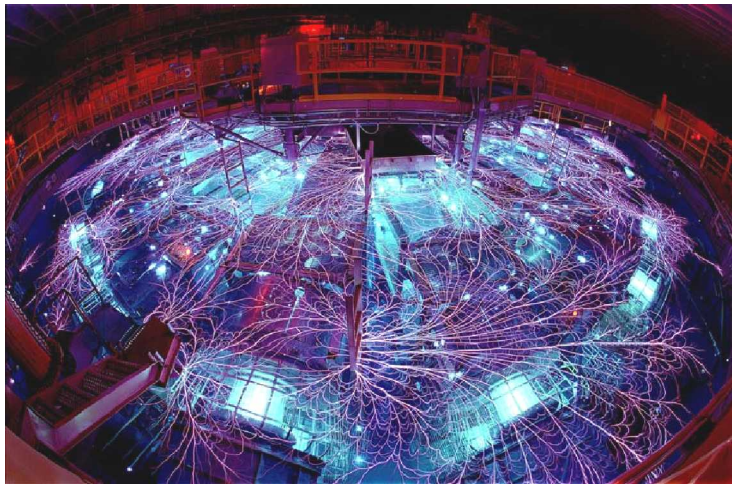
# Motivation for polymer shock simulation

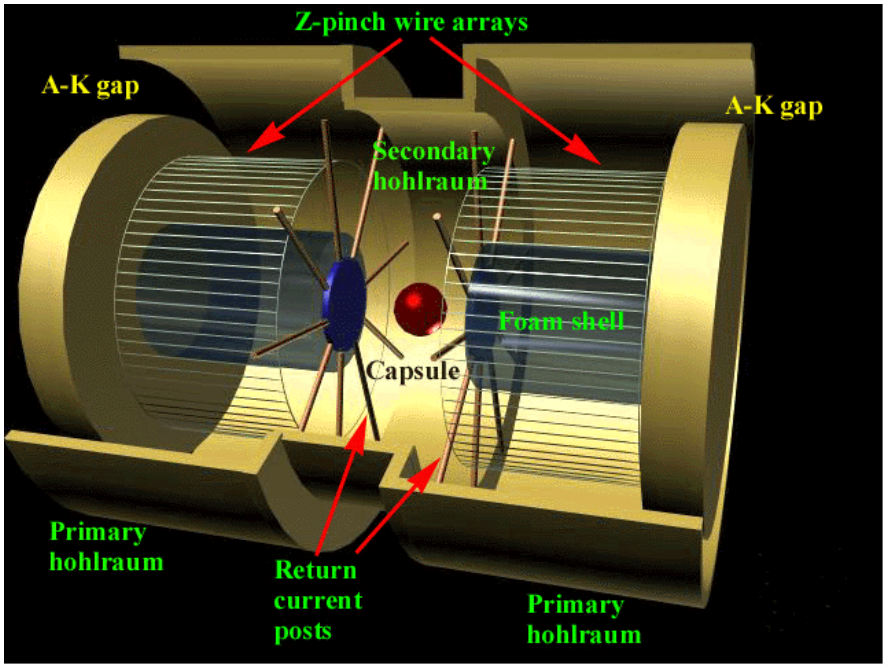
Z-machine at Sandia National Labs





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 foam-shell capsule

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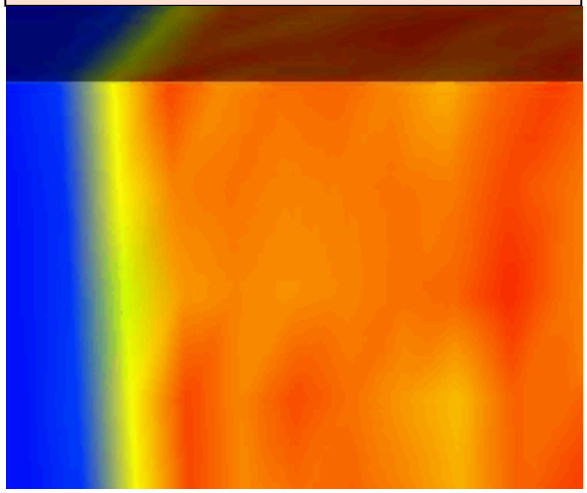
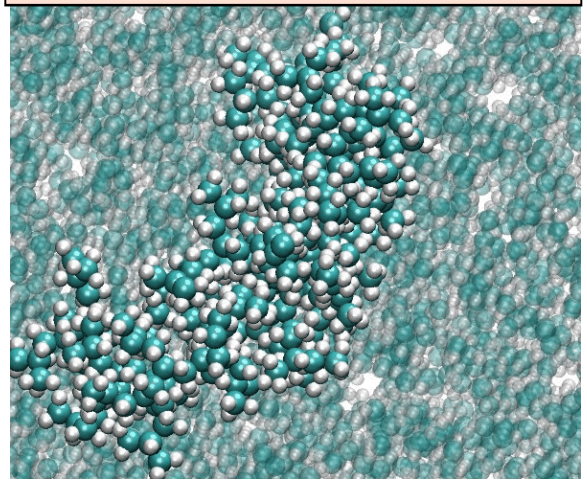
# Hierarchy of modeling approaches

Shock response in hydrocarbon foams depend on processes and structure at several length scales:

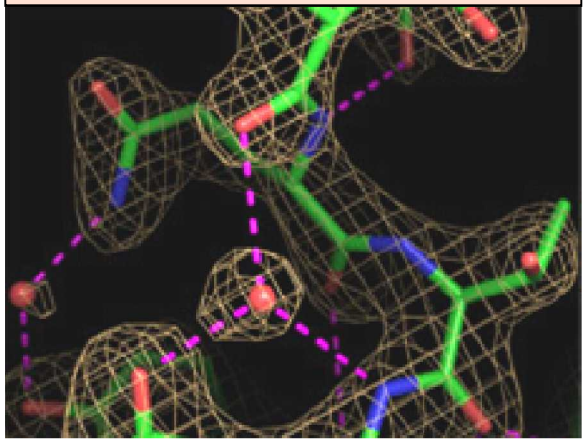
**Continuum scale**
μs times & nm-mm lengths



**Molecular scale**
ns times & 10s nm lengths



**Quantum scale**
fs times & Å lengths



Increasing time and length scales

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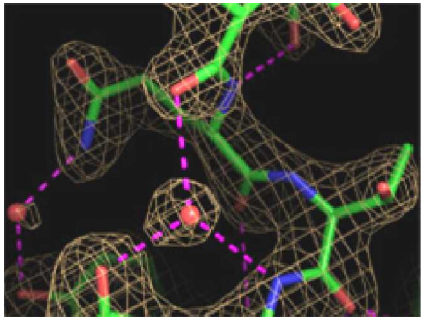
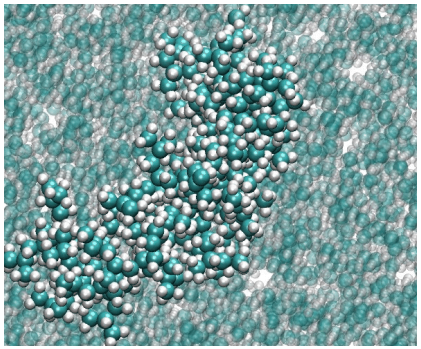
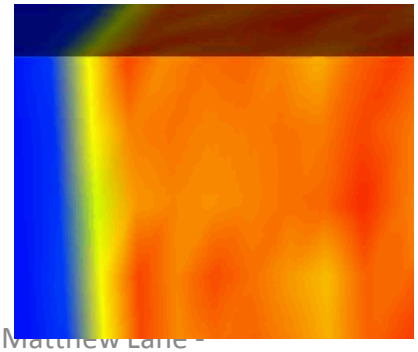
# Hierarchy of modeling approaches



- VASP 5.1 code (Georg Kresse, Vienna, Austria)
- Plane-wave basis 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)
- Cochrane, et al., *Shock Compression in Condensed Matter Proceedings.* (2011)

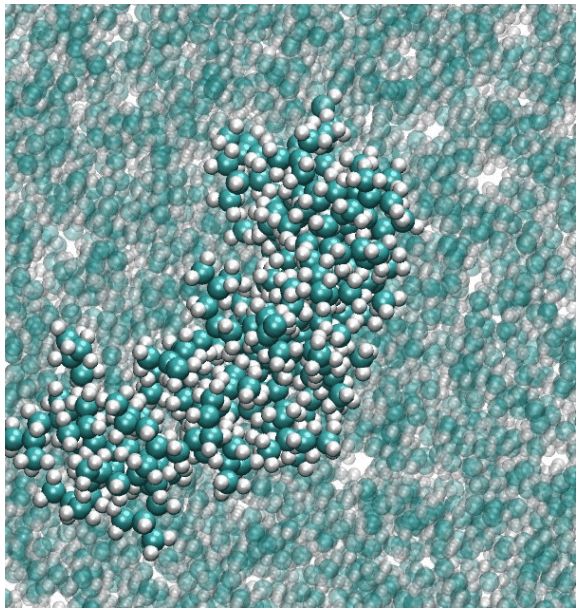


- LAMMPS code (Steve Plimpton, et al., Sandia National Labs)
- MD engine for atoms, molecules, or particles at any length/time scale
- Multiple common interatomic potentials allows for comparison
- Integrated MPI for massively parallel simulations
- http://lammps.sandia.gov
- Lane, et al., *Shock Compression in Condensed Matter Proceedings.* (2011)



- 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
- Haill, et al., *Shock Compression in Condensed Matter Proceedings.* (2011)

# Dense polymer simulation



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

**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 used to compress

**Density Functional Theory**

**AM05** Functional

- Several hundred atom samples of PE and PMP

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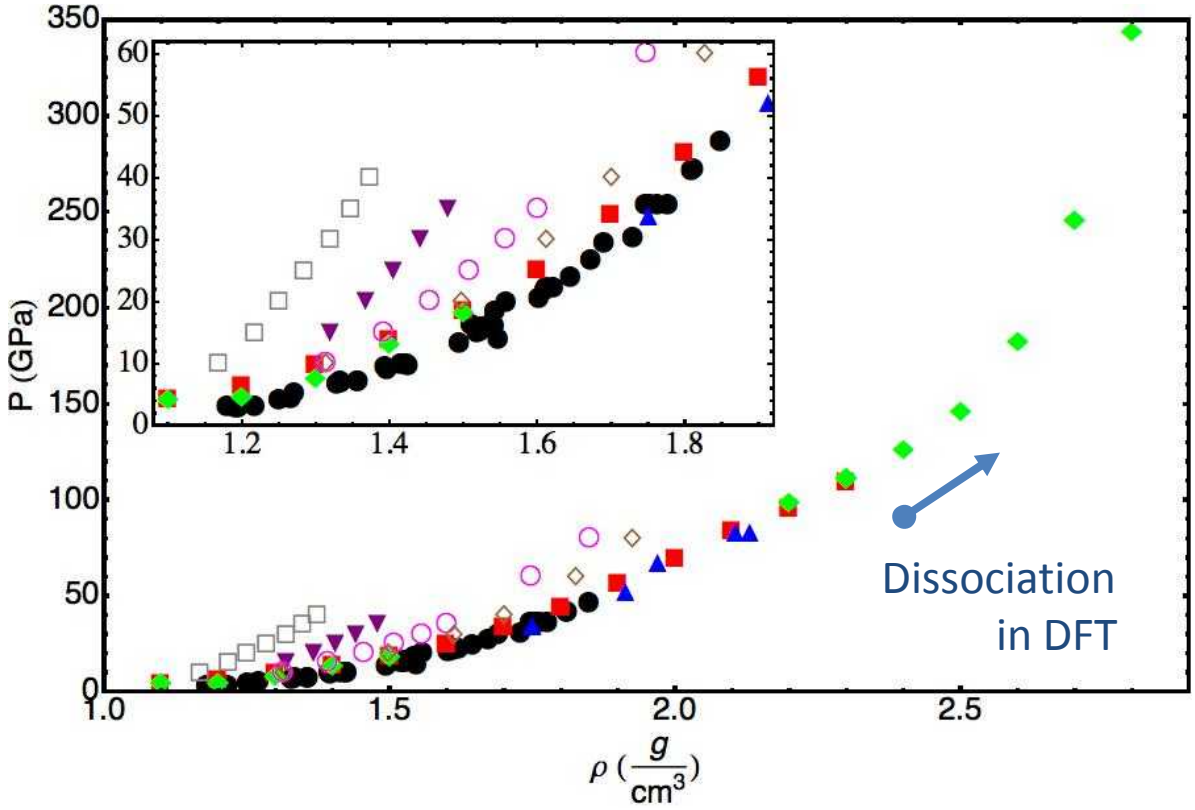
# Polyethylene shock Hugoniot




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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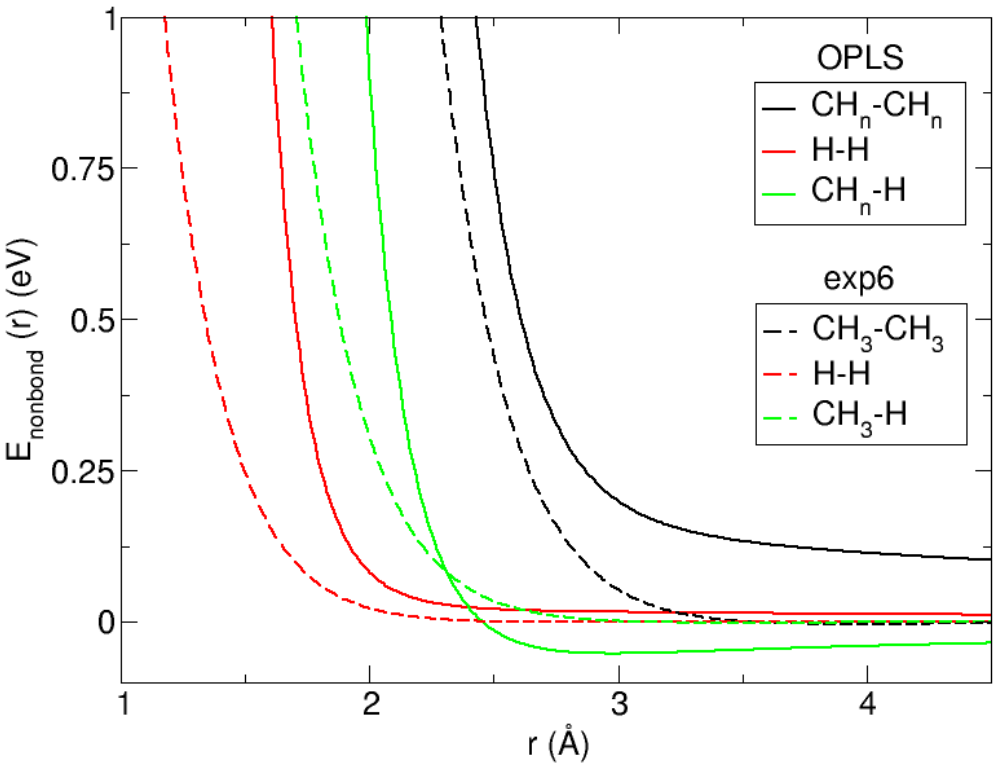
# Core repulsion model is critical




- AIREBO and OPLS share L-J 12-6 functional form for core interaction
- Borodin-Smith exp-6 has a weaker exponential form, more physical shock behavior
- Potentials parameterized near ambient conditions where core interactions are small, unlike in high-density shock conditions
- Behavior under shocks are difficult to predict from equilibrium properties

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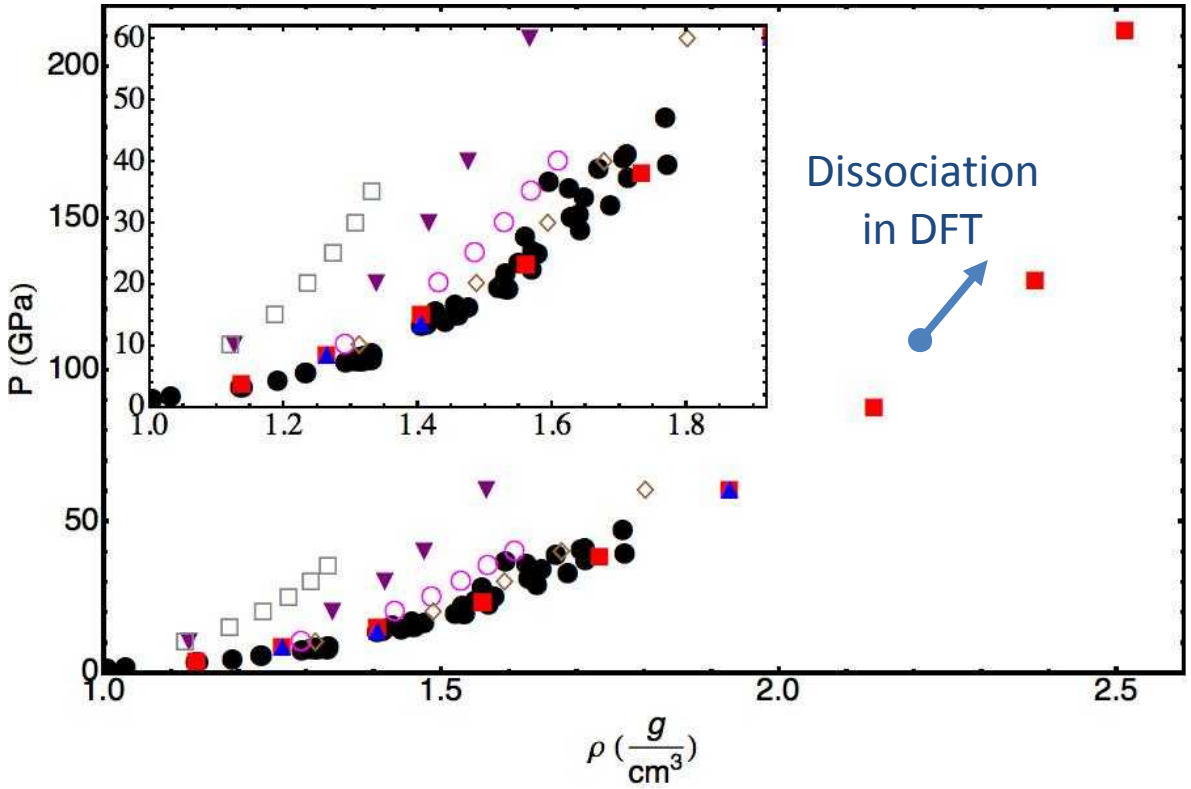
# Poly(4-methyl 1-pentene) shock Hugoniot




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

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

- AIREBO and OPLS both give significantly too stiff shock response at all pressures
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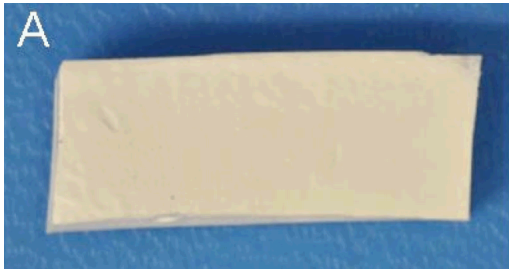
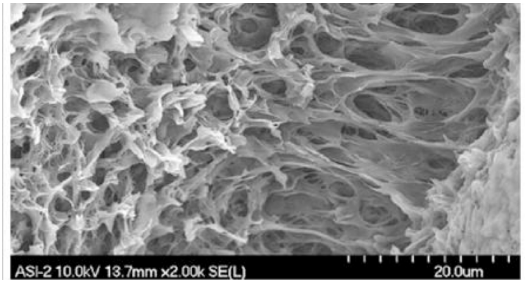
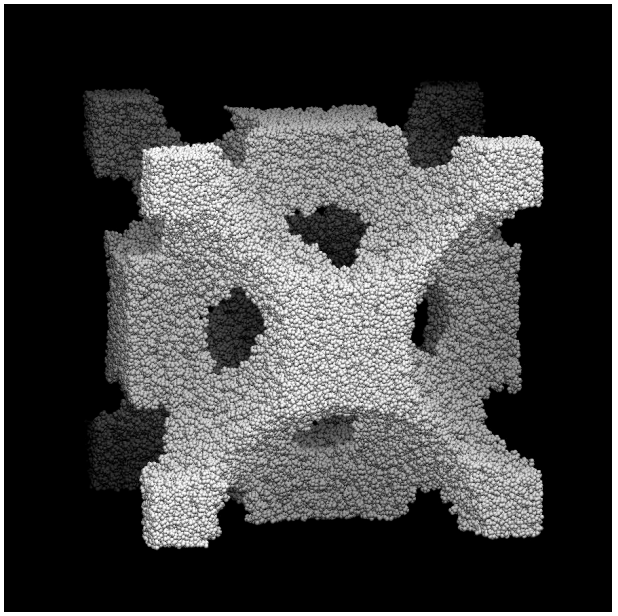
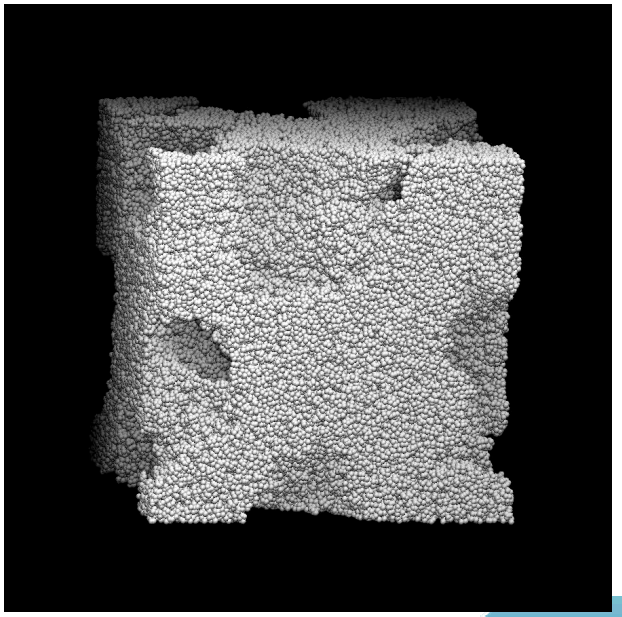
# Foam introduction





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

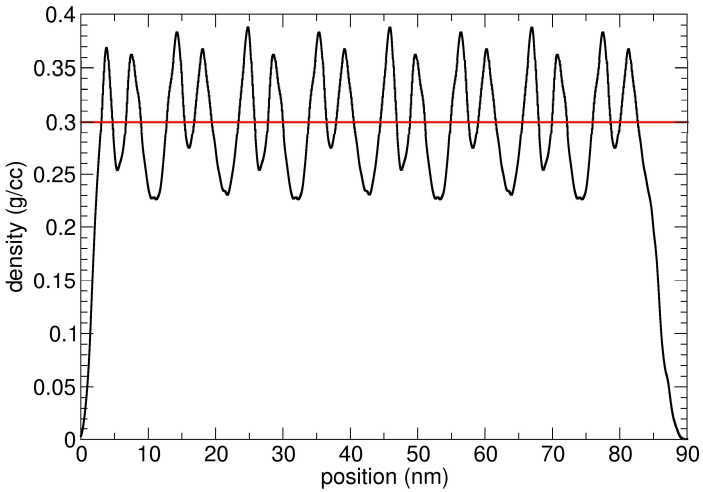
Simulation fabrication:

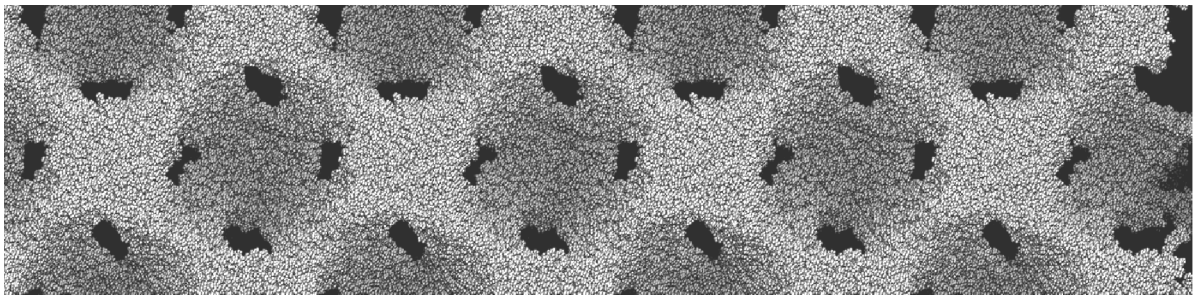
- 0.833 g/cc PMP perforated with growing spherical indenters placed on an fcc lattice, creating a unit cell of foam with unit cell of 20x20x20 nm.

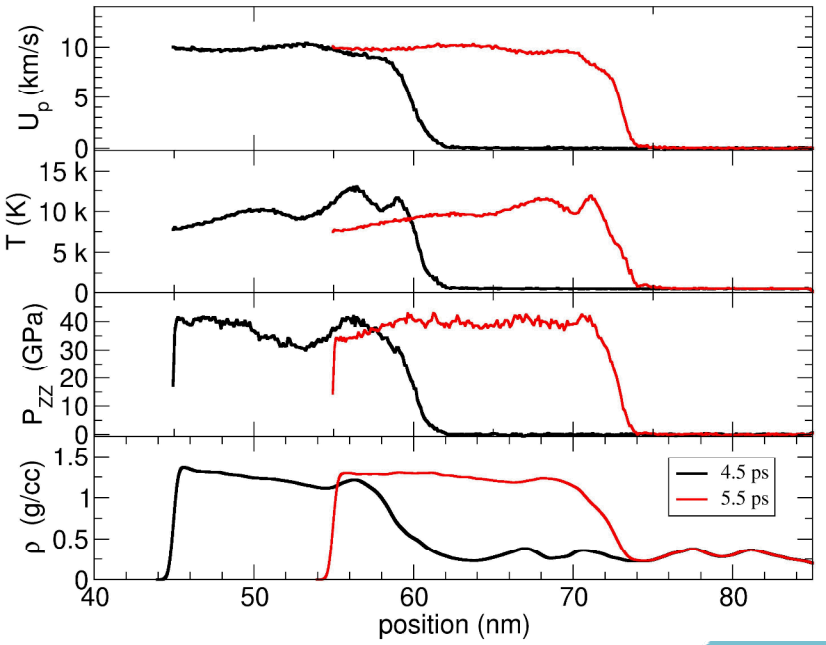
ReaxFF potential was used exclusively.

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

# 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

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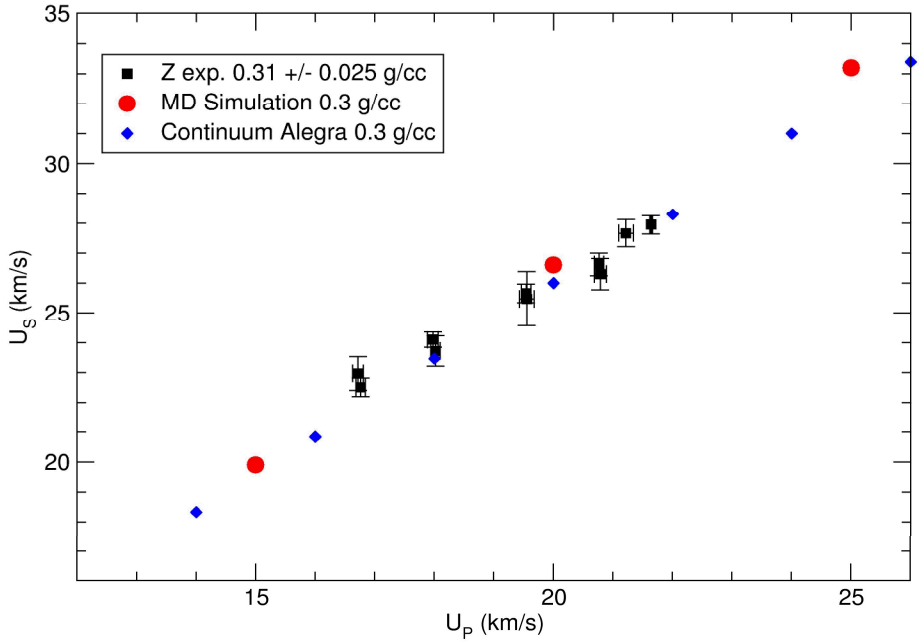
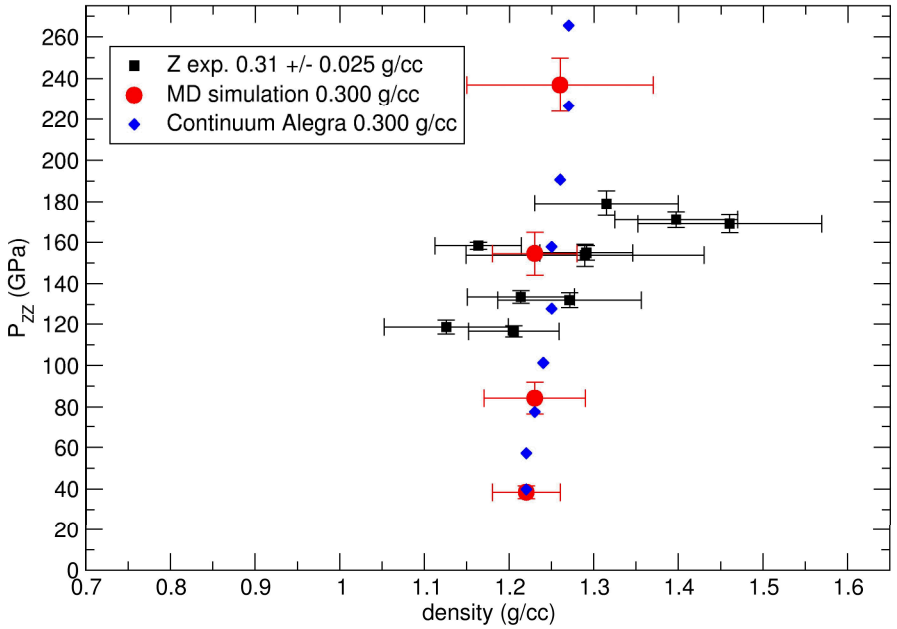
# Foam results – Hugoniot data




- 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.

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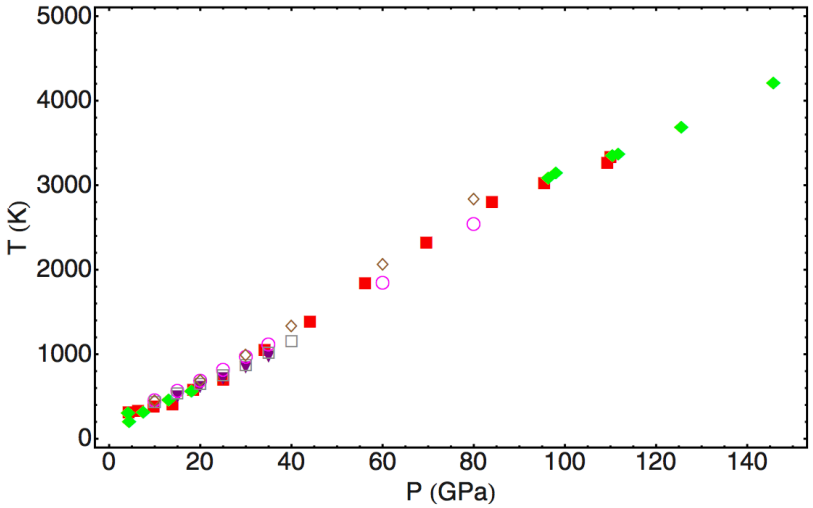
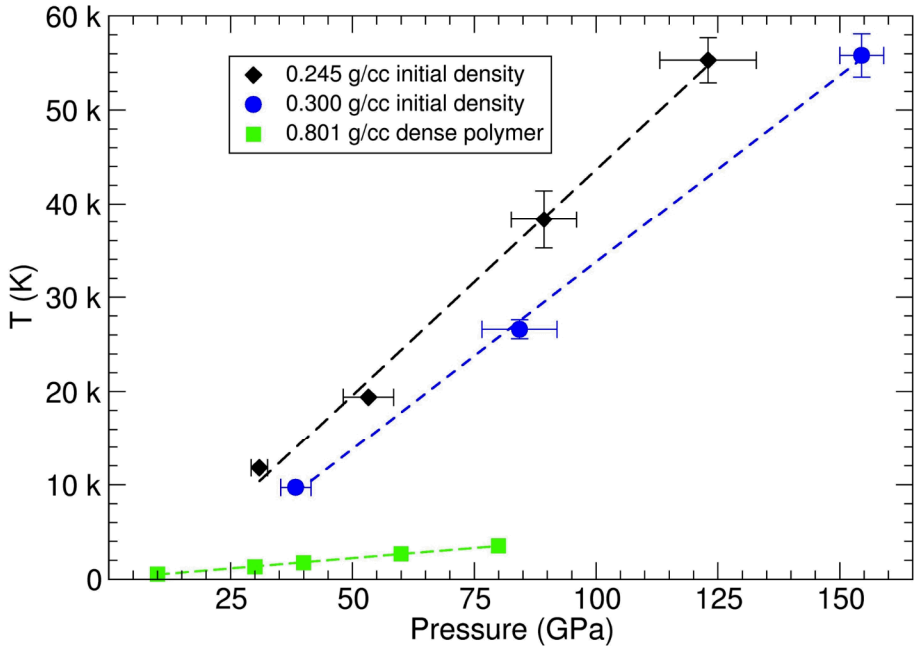
# Temperature dense polymer vs foam




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

Temperatures in dense polymer were < 5000 K through 150 GPA

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

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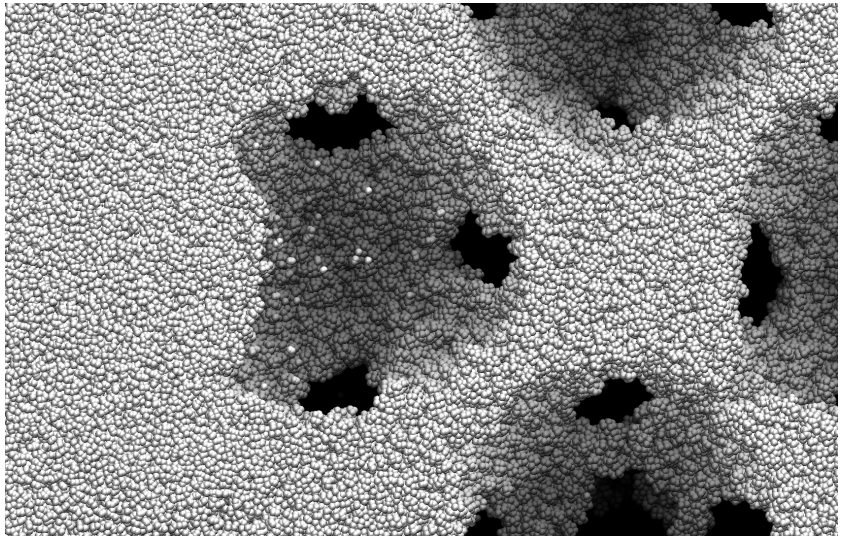
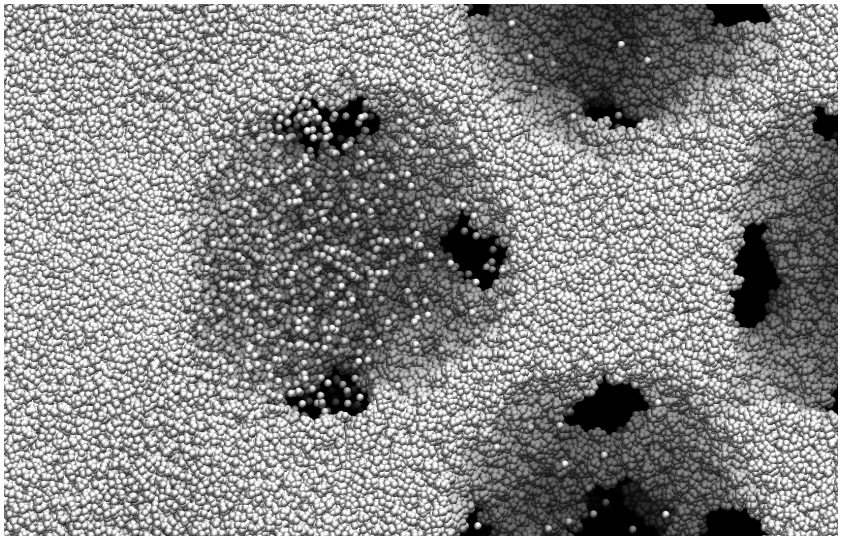
# Foam results – dissociation and vapor

10 km/s piston – 0.300 g/cc PMP foam



25 km/s piston – 0.300 g/cc PMP foam



- Dissociation and vaporization at much lower flyer velocities.
- Accelerated free surface expansion gives rise to void hot spots
- Vaporization and surface blow out broadens the shock profile
- In percolating voids vapor material can behave qualitatively differently than in closed voids.

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# Summary and conclusions

- For dense polymers, **shock response depends more on accurate potentials than on structure**. Thus, DFT is excellent in dense polymers
- **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 found with experiment and continuum simulation for shock compressed polymer foam. Much is left to explore in determining the **role of inhomogeneity, detailed structure and length scales on shock response**

## Future work and possible collaboration opportunities

- Nanoparticle-polymer composites
- Void scale, structure effects and void percolation

## Acknowledgments

Gary Grest, Aidan Thompson, Thomas Haill, Seth Root, Kyle Cochrane, Mike Desjarlais, Thomas Mattsson