SAND2009-6669C

Experiments and multi-scale simulations for shocked polymers and CH_2 foam

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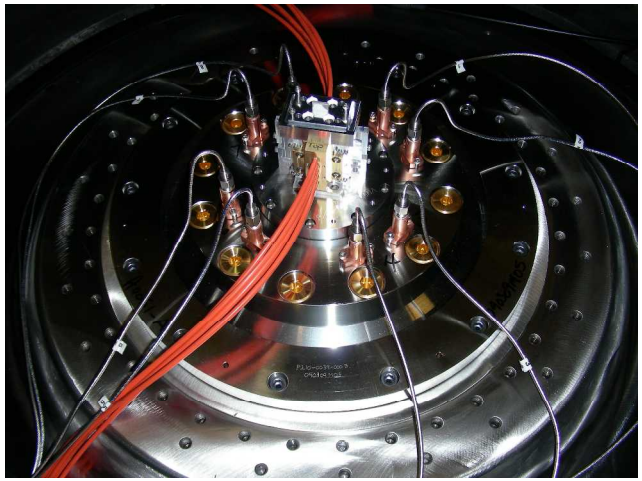
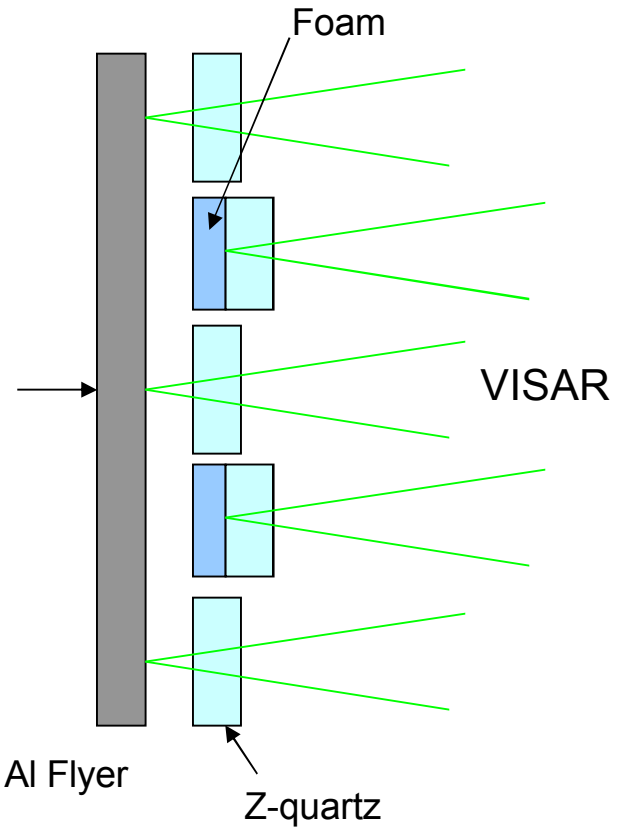
Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the United States Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.

Magnetically accelerated flyer impact experiments

- Four experiments in April 09:
  - 2 – pure, 2 – doped.
- One experiment in August 09:
  - Dense PMP/TPX plastics
- Flyer Velocities:
  - 20.5 km/s to 25.8 km/s



Seth Root experimental PI

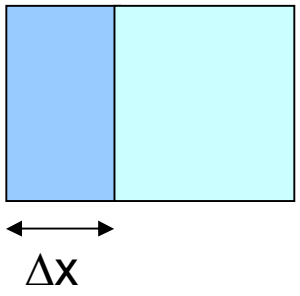


<u>VISAR Measurements as main tool:</u>

- Shock Arrival at Foam/Quartz Interface
- Time of Impact
- Flyer Velocity

Experimental analysis: shock transit time and impedance match with Al standard

- $T_{Transit} = T_{Shock\ Arrival} - T_{Impact}$
- $U_S^{Foam} = \Delta x / T_{Transit}$
- With Al Hugoniot properties, U_S^{Foam}, and ρ_0, we can determine P, U_P, and ρ

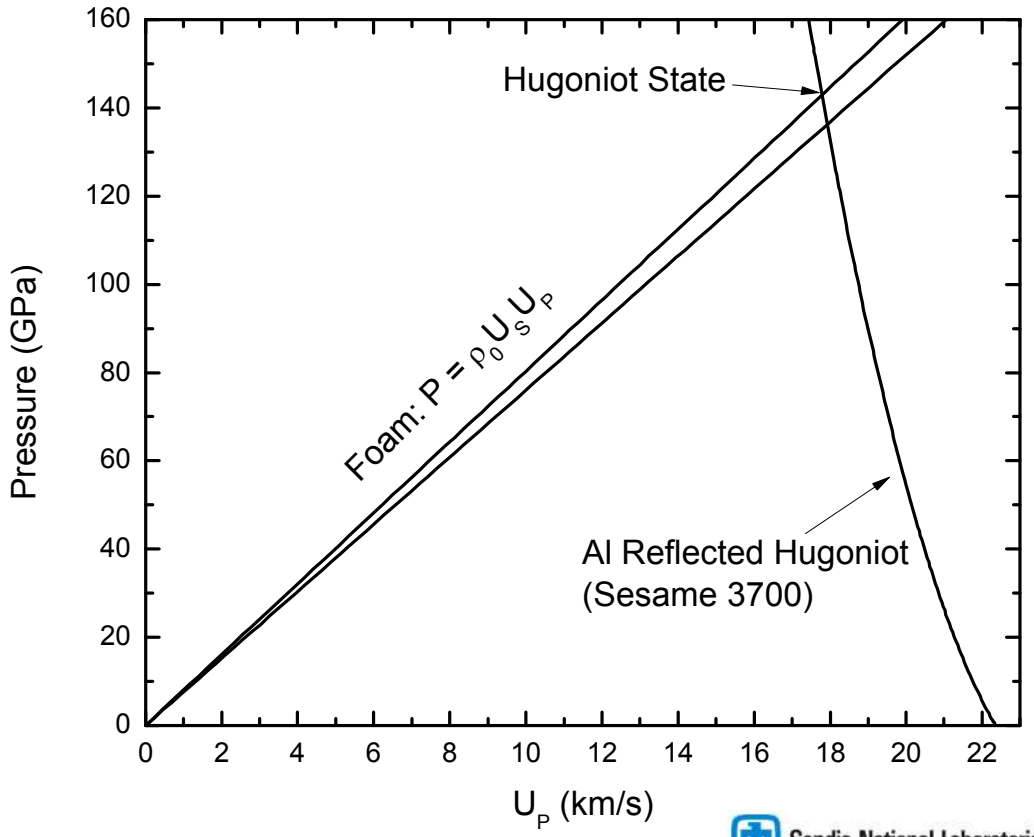


Rankine – Hugoniot Equations:

$P = \rho_0 U_S U_P$

$\rho_0/\rho = (U_S - U_P)/U_S$

$E - E_0 = 0.5(P+P_0)(V_0-V)$



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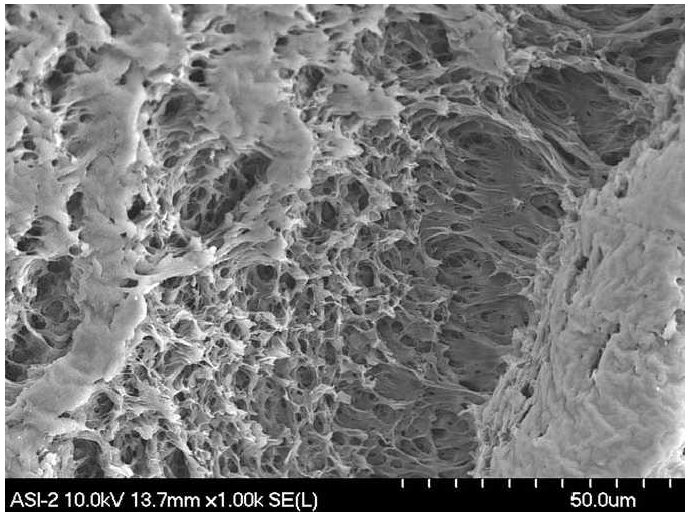
Foam targets made by General Atomics

- Polymethylpentene (PMP/TPX) foam: chemical composition CH_2
- Platinum dopant (150 – 450 nm):
- 50% Pt by weight
- Undoped density = 0.309 g/cm³
- Pt-Doped density = 0.293 g/cm³

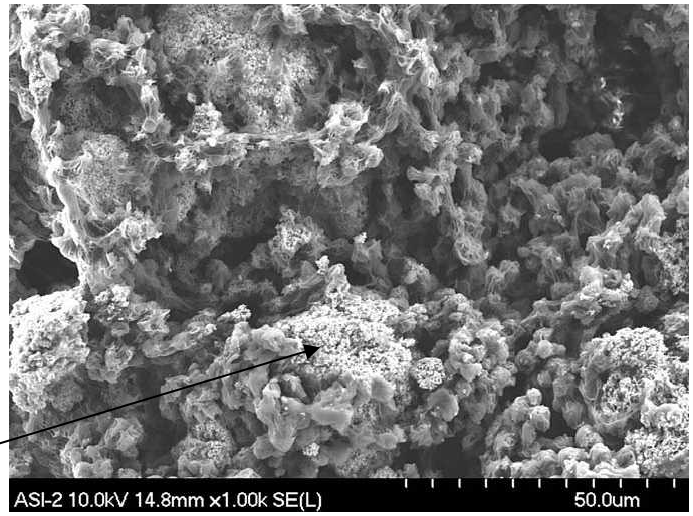
- Targeting 50% mass fraction

Pure Foam

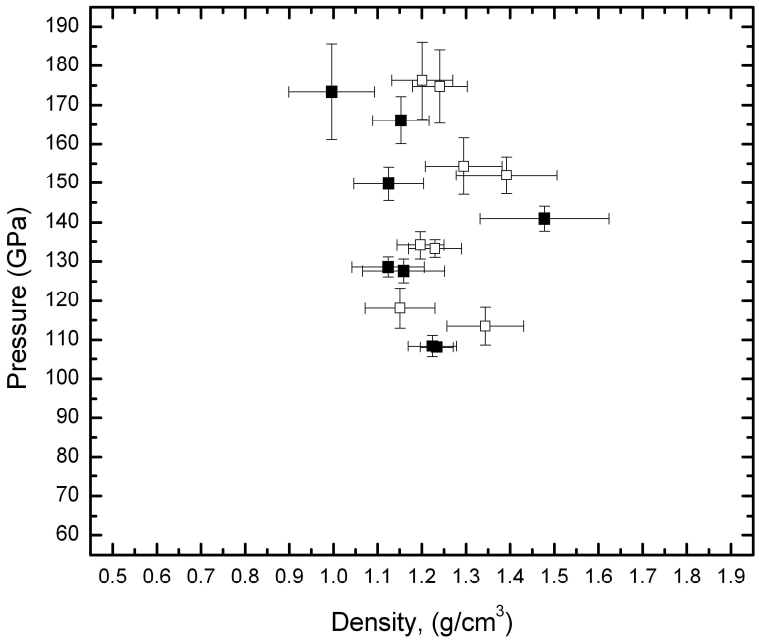


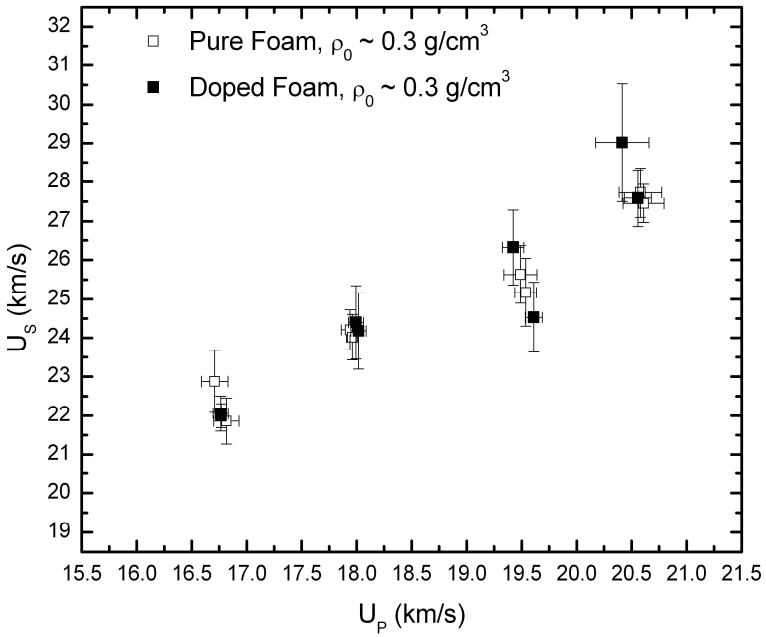
Doped Foam

Platinum



Experimental results: the doped and pure foams show similar response

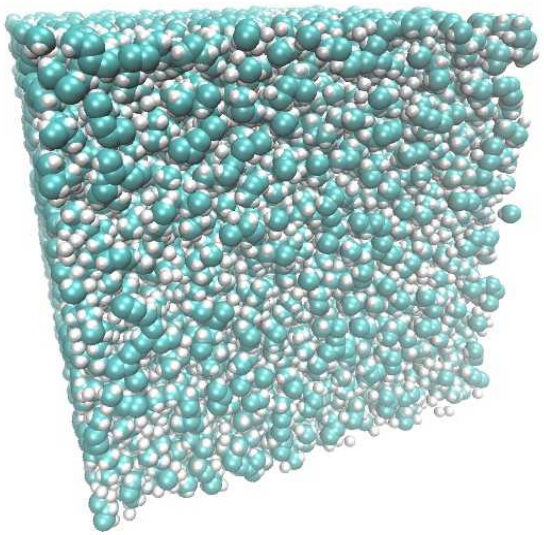




- Pure and doped foam show similar shock response
- Experiments suggest platinum doping at these levels does not affect Hugoniot data

- Error bars are larger than most data from Z
  - Initial density variations
  - Non-uniform shock front

Multi-scale approach to modeling foam, doped foam, and the effects of mixing



Simulation cell for classical MD

- Foam is polymers and void, with the void space collapsing as the foam is shocked
- *Modeling the dense plastic is a key step to modeling the foam*
- Macromolecules are large systems with long timescales, bonded force-fields are commonly used for MD simulations
- *OPLS* (W. L. Jorgensen et al 1996)
- *Borodin-Smith exp-6* (O. Borodin et al 2006)
- Strong shocks will break bonds, consider *reactive force-fields*
- *AIREBO* (S.J. Stuart et al 2000)
- *ReaxFF* (K. Chenoweth et al 2008)
- *Benchmarking is everything. We have done first-principles simulations to investigate the fidelity of these force-fields under shock loading of polyethylene and PMP*

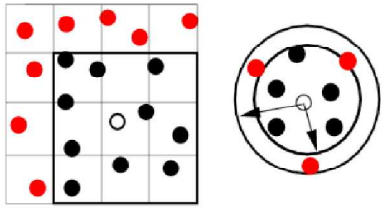
Molecular Dynamics with the Sandia code LAMMPS

What LAMMPS is…

- MD engine for atoms, molecules, or particles at any length/time scale using Newton's equations. Steve Plimpton main developer
- Integrated MPI for parallel or serial for stand-alone *designed for portability and highly efficient parallelization*
- FFT for long-range Coulomb-interactions
- *Many pre-coded potentials:*
  ReaxFF, AIREBO, EAM/MEAM, LJ, Yukawa, Tersoff, etc.
- Open source (GPL)
  http://lammps.sandia.gov

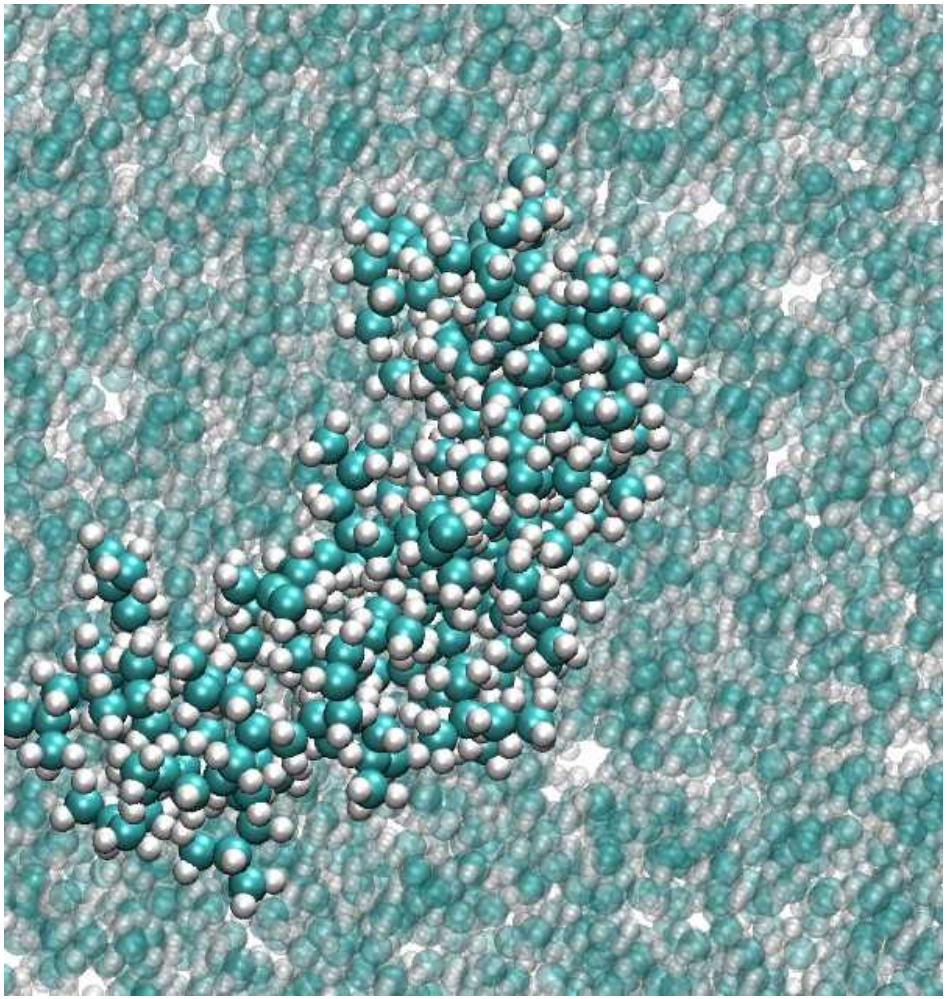
What LAMMPS is NOT…

- Limited pre-processing support for building systems configurations
- Limited on-the-fly diagnostics
- Limited post-processing analysis and visualization
- However, add-on tools and modules are available to accomplish these on website



Computationally efficient neighbor analysis algorithm

Shocked Polymers with LAMMPS



- Four potentials:

  AIREBO, OPLS, ReaxFF, and Borodin-Smith exp-6

- Two polymers:

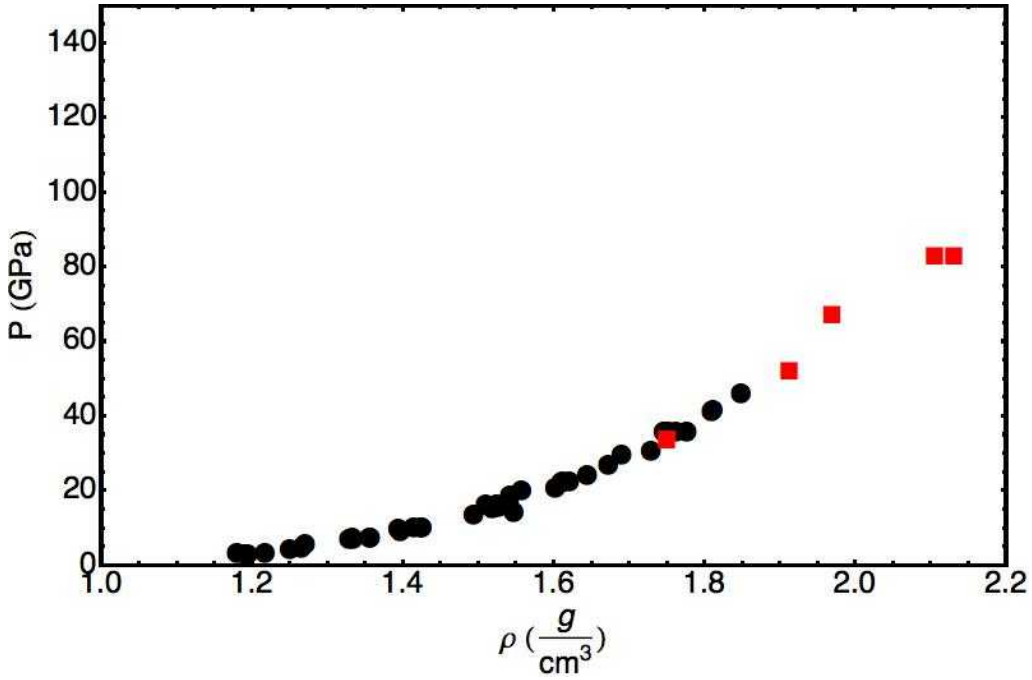
  Polyethylene - simplest possible linear carbon backbone structure forms semi-crystalline solids

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

- 22,000+ atoms in PE sample and 45,000+ atoms in PMP

- Uniaxial Hugoniostat method (Ravelo et al PRB 2004)

Fidelity is everything – treatment of shocked polyethylene can and do vary significantly between models



- LASL shock handbook plus high-pressure work by Nellis and co-workers.

Tight-binding (J.D Kress et al SCCM 1999)
AIREBO
OPLS
Borodin-Smith (exp6)
ReaxxFF
DFT-AM05 (ramp/steady state)
Experiments (Nellis/ LASL handbook)

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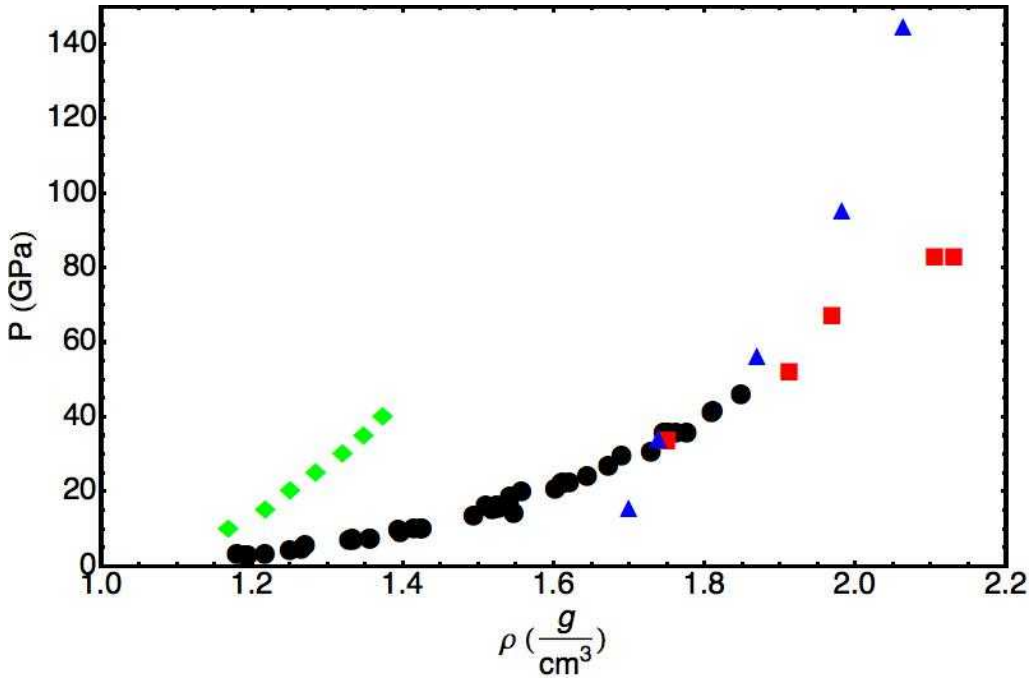

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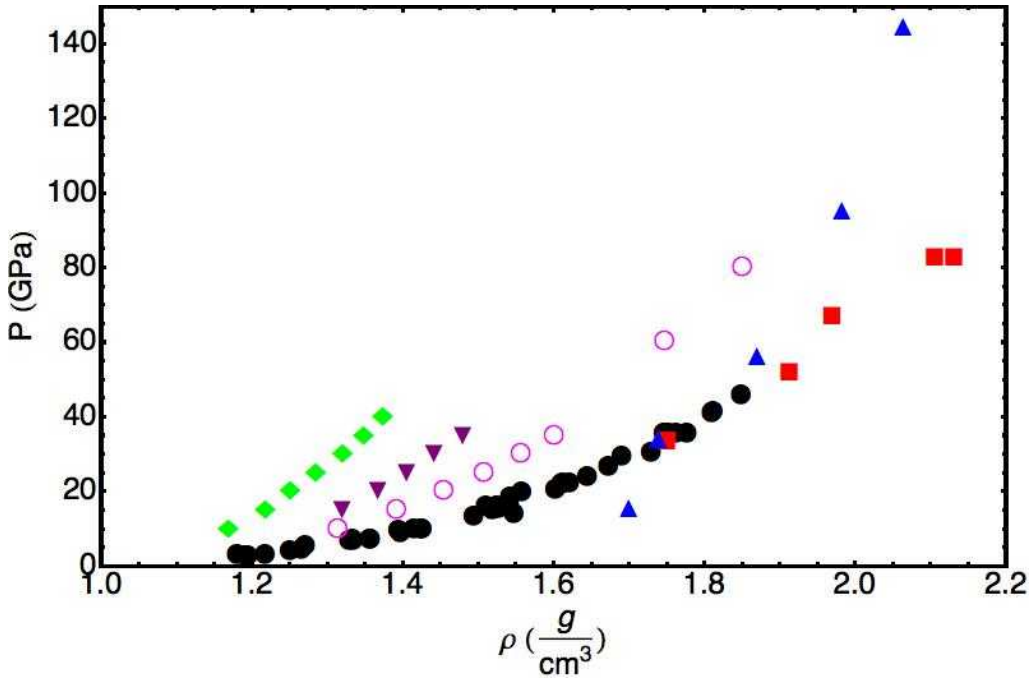

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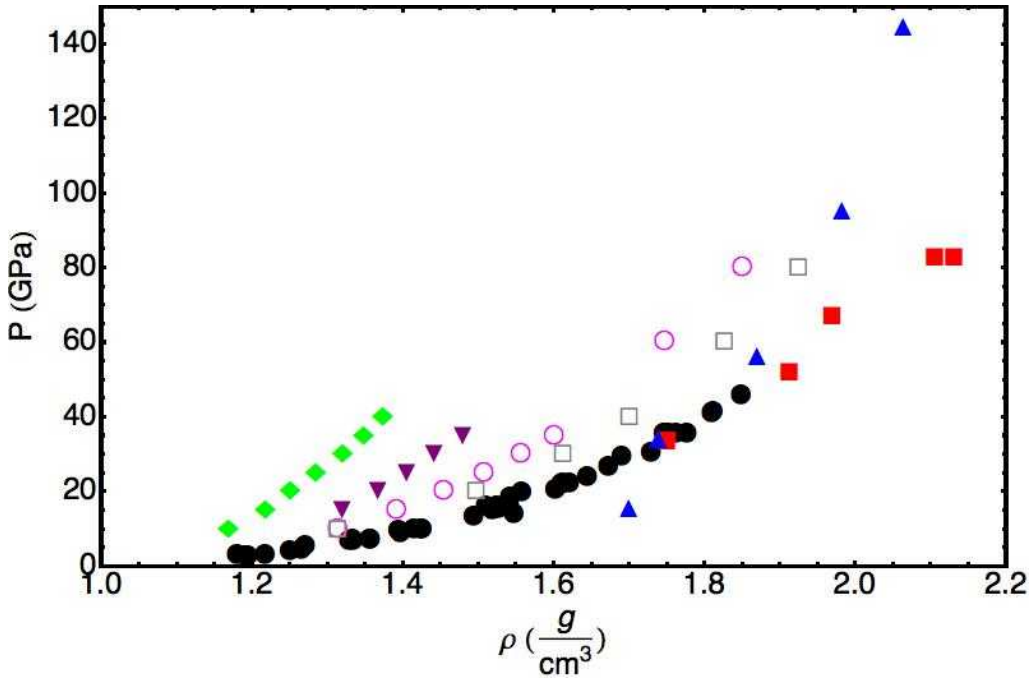

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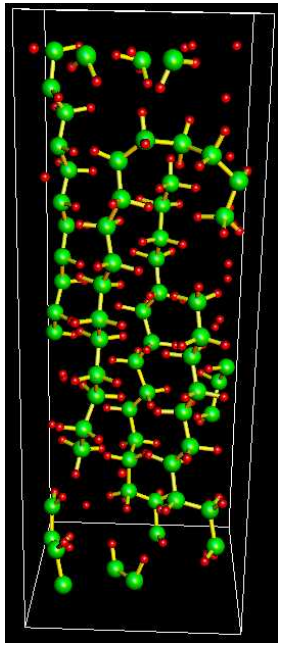
Density functional theory (DFT) based MD is a well-established approach, but far from a black box

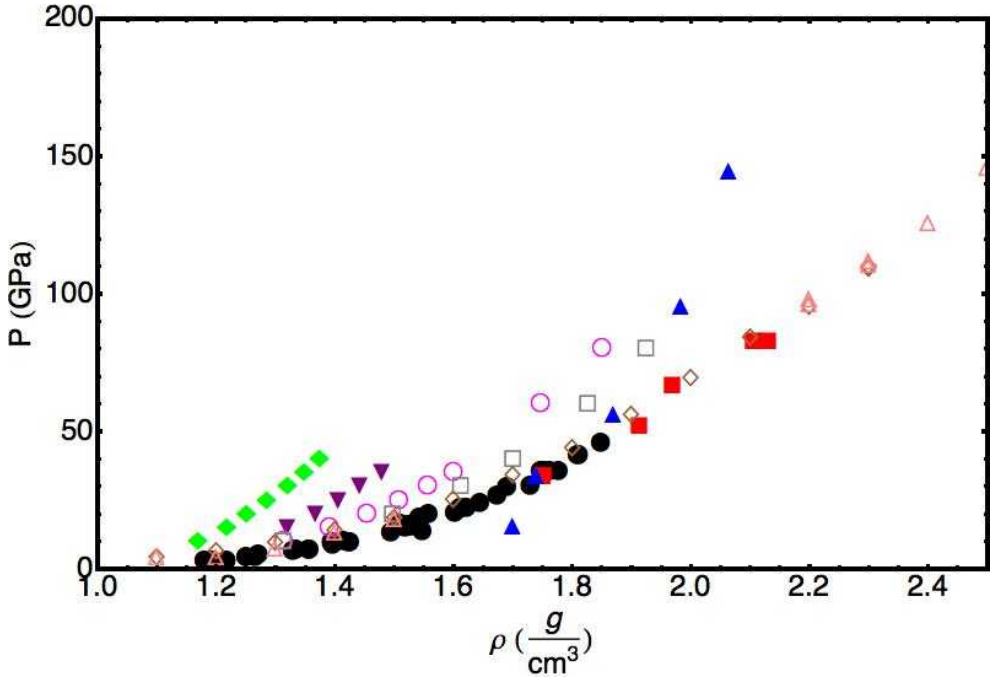


Snapshot of the crystalline polyethylene reference structure at 0.955 g/cm^3 and 300K.

- VASP 5.1 code (Georg Kresse, Vienna, Austria)
- Plane-wave basis-set allowing controlled convergence and free electrons/ionization
- Finite-temperature DFT (Mermin)
- Projector augmented wave core functions (PAW)
- We employ the Sandia developed AM05 functional (Armiento and Mattsson, Phys Rev B 2005
- Accuracy of DFT calculations determined by the exchange-correlation (xc) functional
- Understanding xc functionals/ many-body theory a foundation for high-fidelity simulations: *right answer for the right reason*
- Original research in DFT at Sandia: **dft.sandia.gov** : Rudy Magyar, Anatole von Lilienfeld, Ann Mattsson

- *These are demanding large scale DFT-MD simulations: 200 atoms polyethylene and 440 atoms PMP*
- Utilizing Red Storm at Sandia HPC

Fidelity is everything – treatment of shocked polyethylene can and do vary significantly between models



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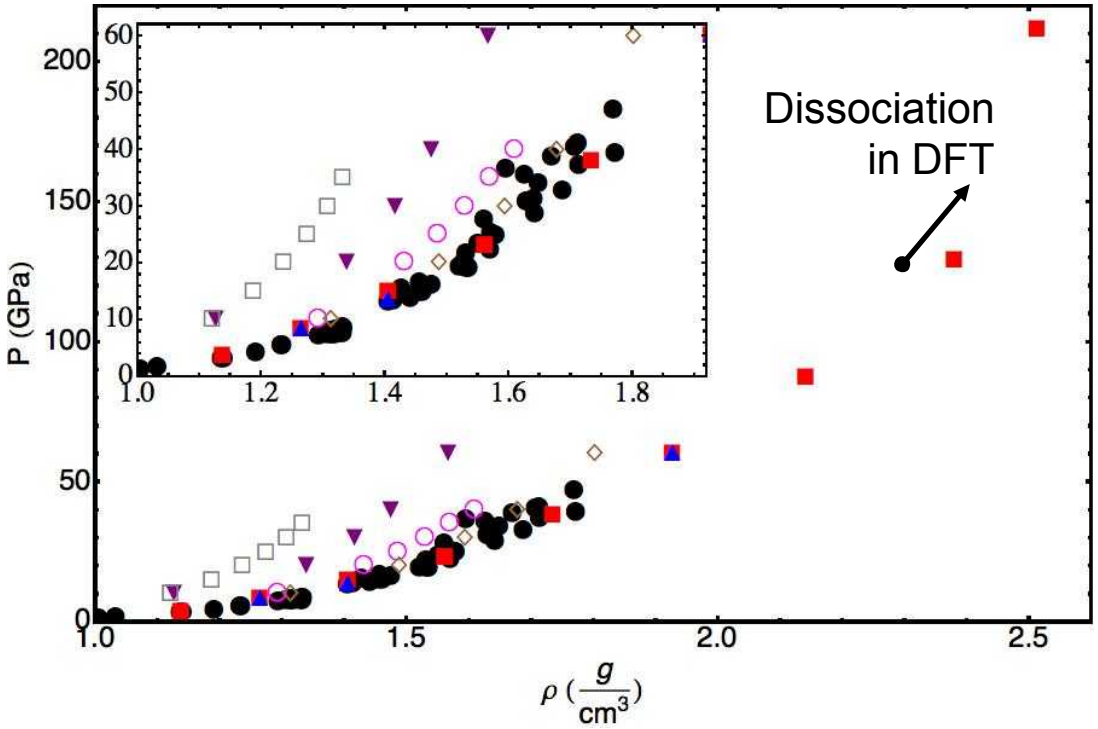
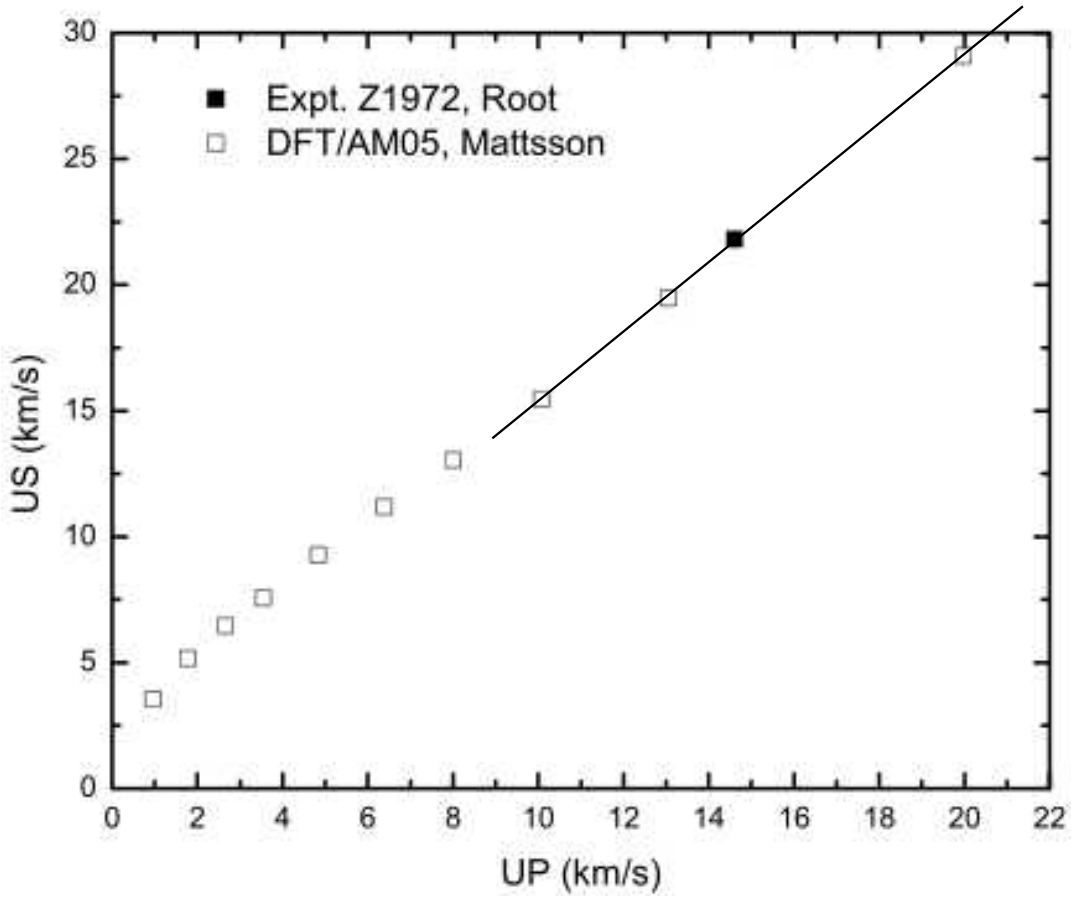
- REAXFF, recent reactive force field, van Duin et al Caltech.
- *DFT-QMD/AM05 in quantitative agreement with shock-data for all compressions.*

Similar differences in behavior for shocked poly(4-methyl 1-pentene)



- AIREBO and OPLS both give *significantly too stiff shock response at all pressures*
- Borodin-Smith and ReaxFF work well for weak shocks in PMP
- *Only the DFT-AM05 simulation of high fidelity for all shocks*
- *Significant deviations already far from the regime where dissociation occurs*
- Classical MD: Gary Grest, Matt Lane, and Aidan Thompson
- DFT-QMD: Polyethylene: Cochrane & Desjarlais, TPX/PMP: Mattsson
- T.R. Mattsson et al, submitted (2009).

Recent Mbar experiments on Z confirms the DFT/AM05 predictions for shocked poly(4-methyl 1-pentene)



- Prediction: DFT-QMD/AM05 simulations using a 440 atom model for PMP
- Flyer plate experiments on Z (Seth Root)
- Excellent agreement between experiments and theory
- *DFT-QMD/AM05 yields results of high fidelity for shocked polymers -- non-trivial finding*
- Gives us confidence in using DFT-QMD /AM05 also for strong shocks in PMP/TPX
- PMP is the polymer from which TPX foam is made

Shocked foam hydrodynamics simulations using the Sandia code ALEGRA

Driver / method

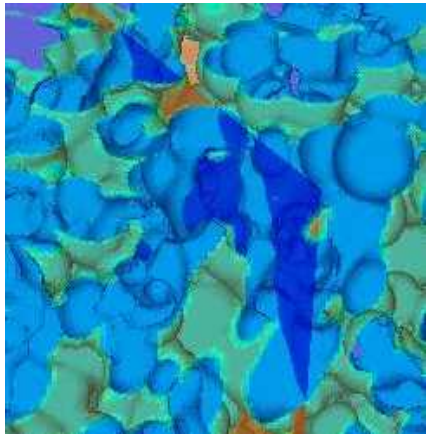
- Constant velocity piston driver
  - Classical shock problem used to generate the Rankine-Hugoniot relationship within the code
- Constant velocity piston driven foam with explicit flyer
  - Reveal issues with driving non-uniform foam with uniform flyer
- Hydrodynamics simulation that includes graded density/temperature flyer at time of impact
  - Initial flyer conditions from MHD simulation of flyer only
- Full MHD drive simulation of flyer / foam / window
  - Future
- Lagrangian simulations

Target material models

- Homogenous low-density plastic
  - Initialize uniform material at average foam density
- Porosity homogenous target
  - Invoke the P-alpha model
  - Specify solid plastic density as reference
  - "$\alpha = \rho_{solid} / \rho$" is the distention parameter reduces to 1.0 as void compressed from foam
- Mesoscale direct simulation of foam
  - Explicitly model plastic matrix with voids
  - Development state
- Mix models
  - Volume fractions
  - Isentropic mix

Porosity is modelled in ALEGRA using the P-α model

- The distention parameter α is defined and advanced in time
  - $\alpha = \rho_{solid} / \rho_{foam} \geq 1$
  - Evolution equation for α
- Pressure and energy/temperature tables are evaluated at the compressed solid density when $\alpha > 1$
  - $P(\rho_{foam}, T_{foam}) = 1/\alpha\ P_{table}(\alpha^*\rho_{foam}, T_{foam}) = 1/\alpha\ P_{table}(\rho_{solid}, T_{foam})$
  - $E(\rho_{foam}, T_{foam}) = E_{table}(\alpha^*\rho_{foam}, T_{foam}) = E_{table}(\rho_{solid}, T_{foam})$
  - Otherwise tables are evaluated normally when $\rho > \rho_{solid}$ (i.e., $\alpha = 1$)
- References:
  - W. Herrmann, J. Appl. Phys., 40 (6) 2490, May 1969.
  - M.M. Carroll and A.C. Holt, J. Appl. Phys, 43 (4) 1626, April 1972.
  - G.I. Kerley, SAND92-0553, SNL, April 1992.

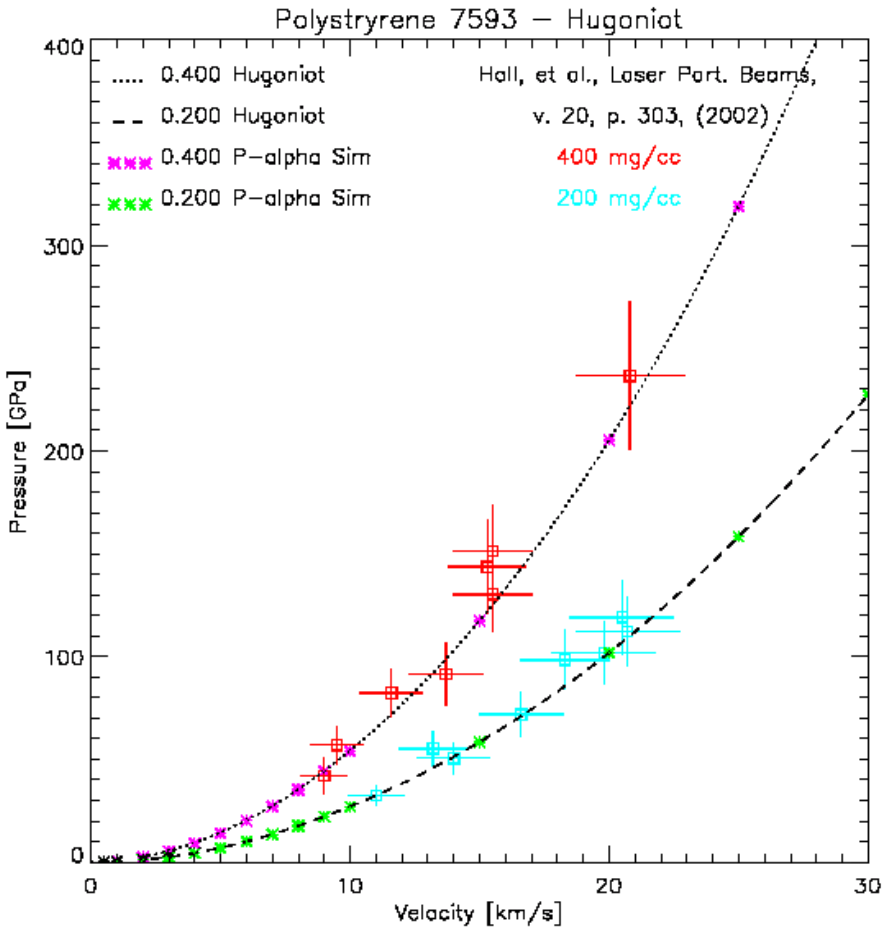


- Pressure reduced by distention parameter since a porous material can't sustain the pressure of the bulk material
- Energy from bulk material (neglect surface effects) questionable assumption in high surface/volume materials like foam
- *Must validate the model*

Validation of P-α Model for Polystyrene

- Partial Hugoniot data in literature for TMPTA foam
- Polystyrene is a close approximation
  - Hall, et al., used 7592
  - We use Aneos 7593
    - » More complete
    - » More consistent
- Laser driven shock experiments on LULI, France
- ρ = 20, 50, 100, 200, 400, and 1100 mg/cm^3
- We model ρ = 200 and 400 mg/cm^3

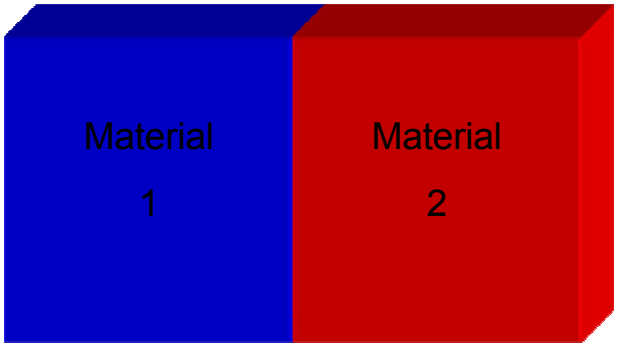
- *Data and simulations agree without tuning, initial density is not a free parameter*



EOS Mixing Rules (aka Multi-Material Treatment) in ALEGRA

- Whenever a cell compresses or expands, how do the volume fractions change?
- This affects densities and specific internal energy
- Hence also affects temperature and pressure

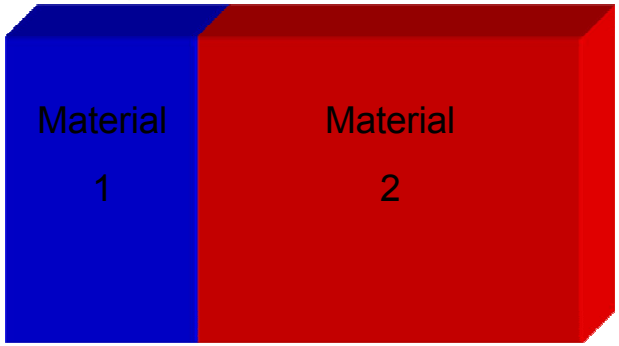
- Default (legacy) method:
  - Volume fractions, f_1 and f_2 , held constant
  - Density & energy adjusted for each material separately
    - » $\rho_{new} = \rho_{old} * (V_{old} / V_{new})$
    - » $e_{new} = e_{old} * (\rho_{old} V_{old} / \rho_{new} V_{new})$
  - Temperature & pressure updated by EOS evaluation



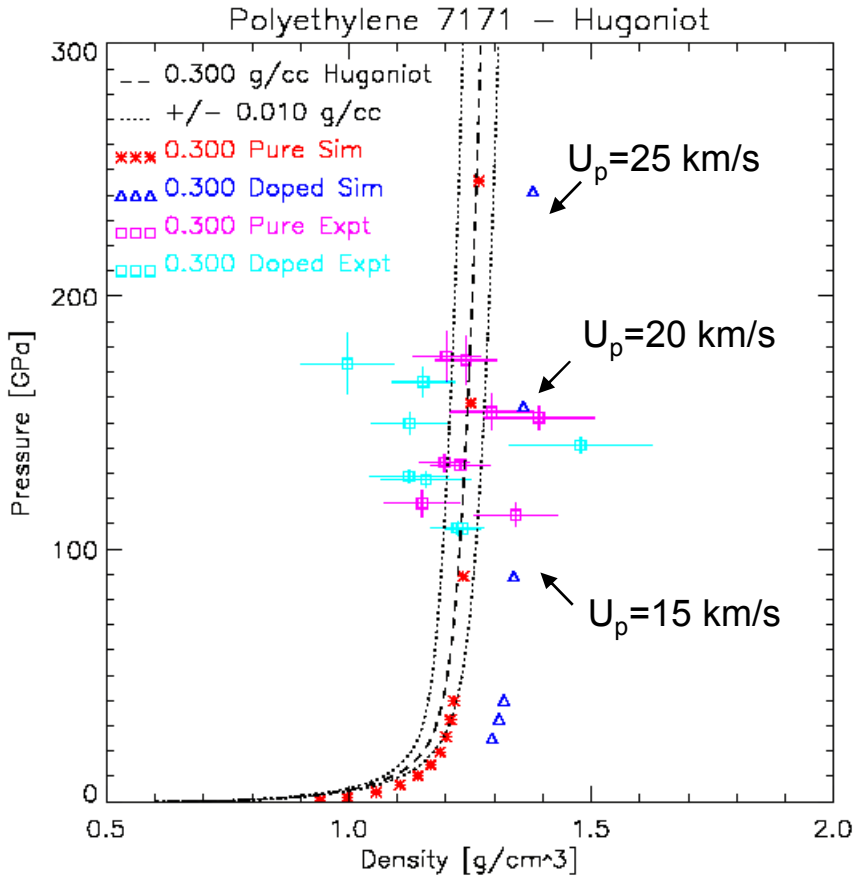
- Isentropic Compression Method (new):
  - Update volume fraction for each material *k* using bulk modulus B_k

$$\frac{df_k}{dt} = f_k \left(\frac{\overline{B} - B_k}{B_k} \right) \nabla \bullet \vec{u} - \frac{f_k}{\overline{P}} \frac{dP_k}{dt}$$

- Normalize volume fractions to 1.0
- Enforce mass & energy conservation
- Update EOS

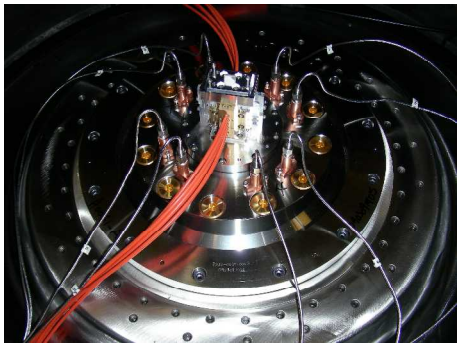


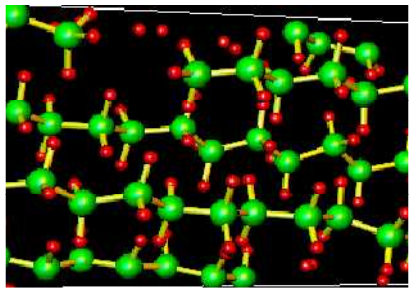
Homogenous pure and platinum-doped foam simulations shock to same pressures and agree with Z experiments

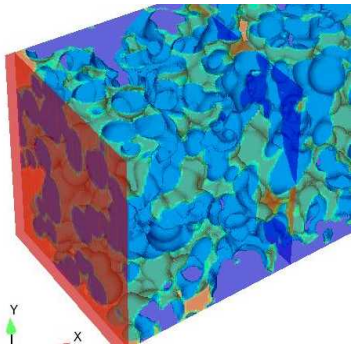


- Pure and doped foams nominally 0.300 g/cm^3 average density (experimental 0.29 and 0.30 g/cm^3)
- Three lines are pure foam at 0.29, 0.30, 0.31 g/cm^3, respectively.
- Platinum-doped foam is 50-50 mixture by weight
  - *99.3% foam and 0.7% platinum by volume*
- Simulations use P-α model for foams
- Doped foams shock to same pressures at pure foams

Future experimental and theoretical work on foam to form a comprehensive understanding of the problem



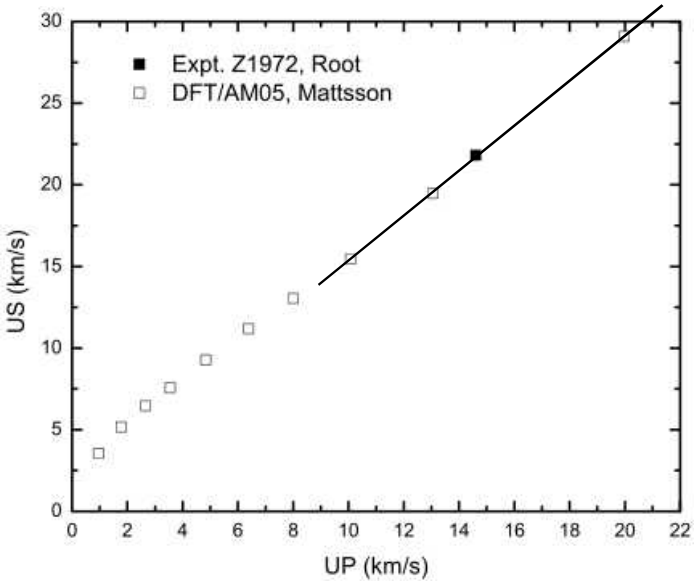




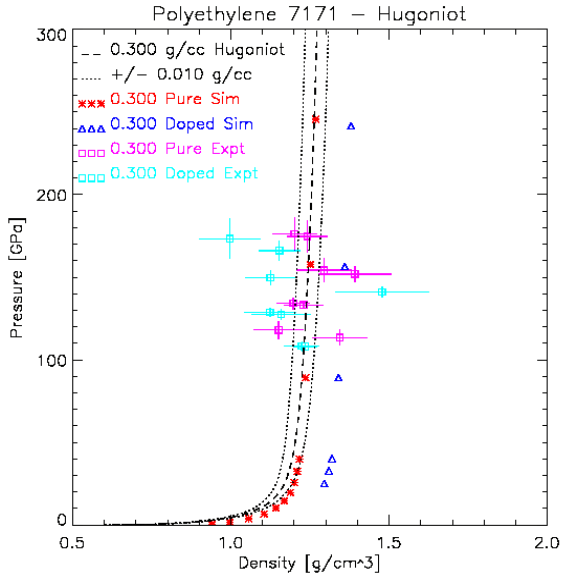
- Expand Hugoniot measurements on pure and doped foams with ρ ~ 300 mg/cc
- Compare 300 mg/cc pure foam with 600 mg/cc doped foam (300 mg/cc of foam with 300 mg/cc of Pt) (*is being analyzed).*
- *Measure the Hugoniot for the dense TPX plastics, to validate the DFT/QMD simulations (did this in August)*
- Ultra-high flyer velocities (40+ km/s)

- Large-scale classical MD for porous properties
- DFT/QMD simulations on high-pressure response of foams
- Continued MHD simulations with meso-scale structure
- Comparisons with mix rules / multi-material treatment

Conclusions and summary



DFT-QMD/AM05 predictions for high-pressure Hugoniot from validated by recent experiments on Z.



Promising results for CH2 foam as a system to experimentally validate models for mixed equation of state

Tom Haill, Gary Grest, Matt Lane, Aidan Thompson, Kyle Cochrane, Michael Desjarlais, Seth Root, Ray Lemke, Dawn Flicker, Tom Mehlhorn.

Backup slides to follow

Shock waves

Shocked foam applications

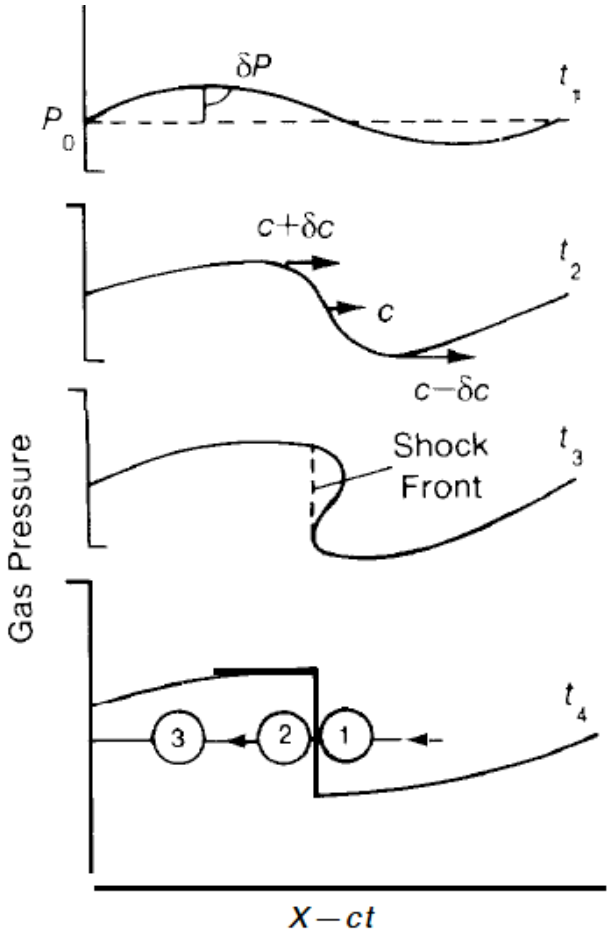
DFT-XC AM05 solids benchmark

P-a model more validation

Piston driven foam calculations

More on nano voids and particles/clusters

Shock waves: discontinuity sets a thermodynamic constraint on the state after the shock wave

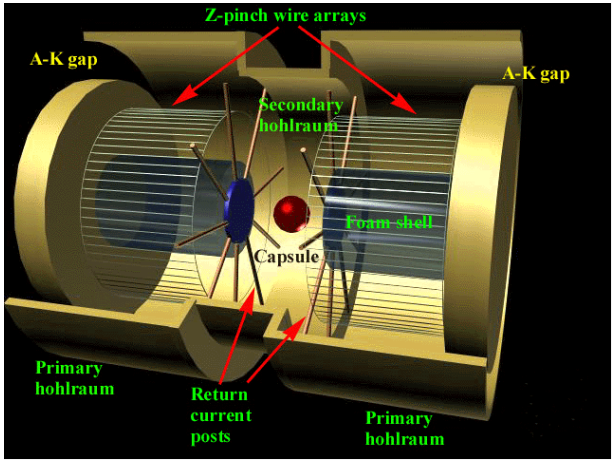


- Density dependence of wave velocity
- Discontinuity at shock, but a steady traveling wave
- $V_{shock} > V_{sound}$
- Conservation of mass, energy, and momentum lead to the Rankine-Hugoniot condition for the initial (1) and final state (2)

$$2(U_2 - U_1) = (P_2 - P_1)(\upsilon_2 + \upsilon_1)$$

- U - internal energy
- P - pressure
- v - volume
- *Valid above the yield strength of the material*

Properties of shocked hydrocarbon foams is of importance to modeling inertial confinement fusion



Double-ended Z pinch with an ICF foam shell capsule

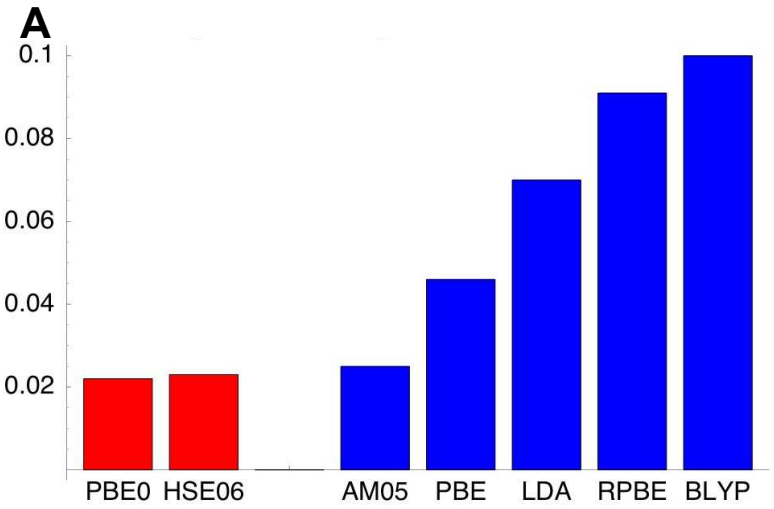
Shock experiments reach conditions of giant planets like Jupiter and Neptune

- A commonly used foam is based on poly(4-methyl 1-pentene) (PMP/TPX)
- Modeling the dense plastic as a step to modeling the foam
- Macromolecules are large systems with long timescales, bonded force-fields are commonly used for MD simulations
- *OPLS* (W. L. Jorgensen et al 1996)
- *Borodin-Smith exp-6* (O. Borodin et al 2006)
- Strong shocks will break bonds, consider reactive force-fields
- *AIREBO* (S.J. Stuart et al 2000)
- *ReaxxFF* (K. Chenoweth et al 2008)
- *Benchmarking is everything. Sandia did first-principles simulations to investigate the fidelity of these force-fields under shock loading of polyethylene and PMP*

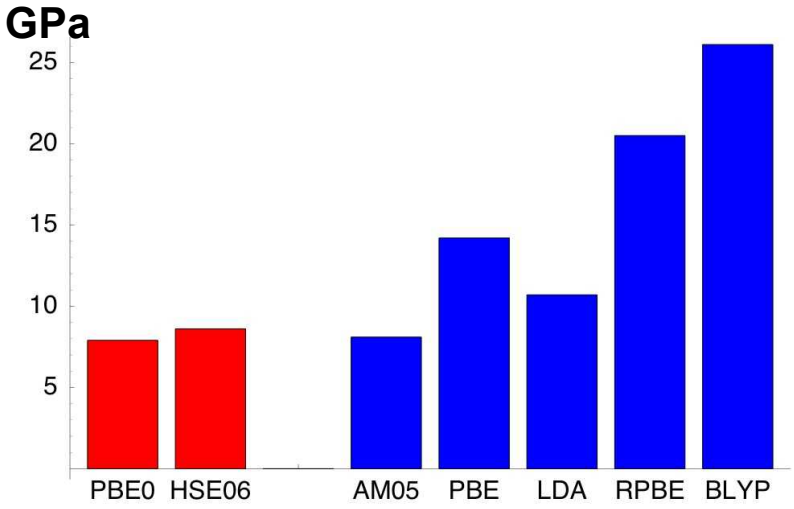
A new level of accuracy using the new XC functional AM05: benchmark for 20 solids[$]

Li, Na, Al,
BN, BP, C, Si, SiC,
β-GaN, GaP, GaAs,
LiF, LiCl, NaF, NaCl, MgO,
Cu, Rh, Pd, Ag.

AM05 performs as well for solids as the hybrids HSE and PBE0 do, while being 60-1000 times less expensive in computer time.
Accurate and fast is possible



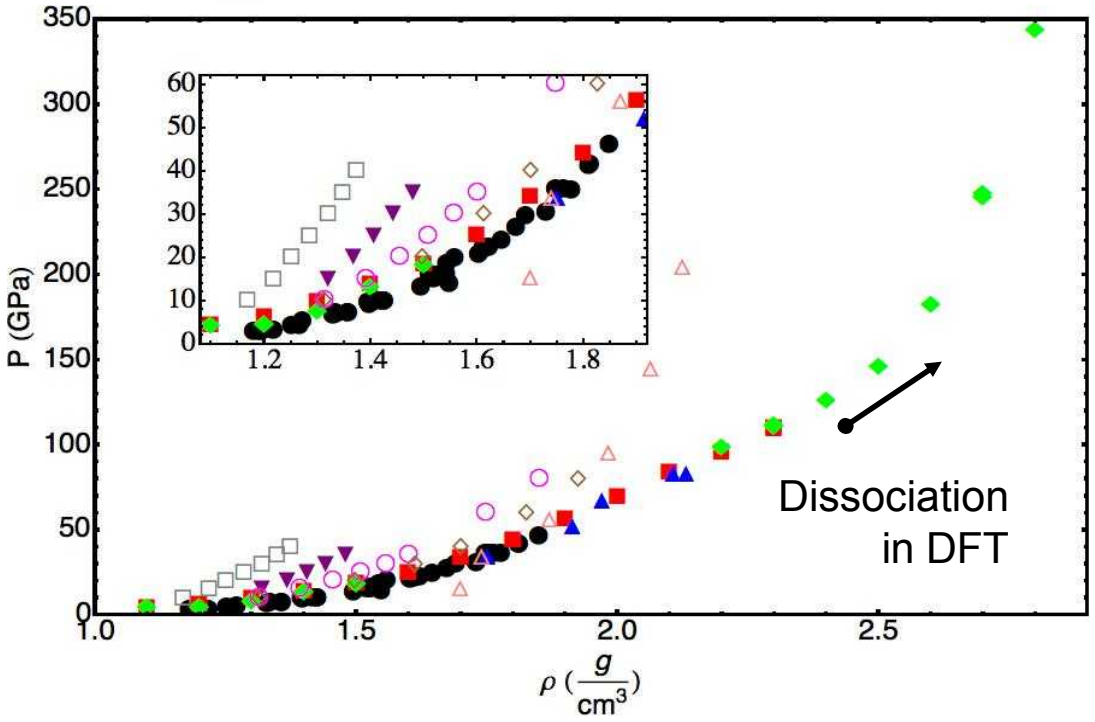
Mean absolute error: lattice const.



Mean absolute error: bulk moduli.

[$]A.E. Mattsson, R Armiento, J Paier, G. Kresse, J.M. Wills, and T.R. Mattsson, J. Chem. Phys. **128**, 084714 (2008).

Qualitative differences in behavior for shocked polyethylene between different force-fields



- Tight-binding (J.D Kress et al SCCM 1999)
- AIREBO
- OPLS
- Borodin-Smith (exp6)
- ReaxxFF
- DFT-AM05 (ramp/steady state)
- Experiments (Nellis/ LASL handbook)

- 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*
- Classical MD: Gary Grest, Matt Lane, and Aidan Thompson
- DFT: Polyethylene: Cochrane & Desjarlais, TPX/PMP: Mattsson

- T.R. Mattsson et al, submitted (2009).

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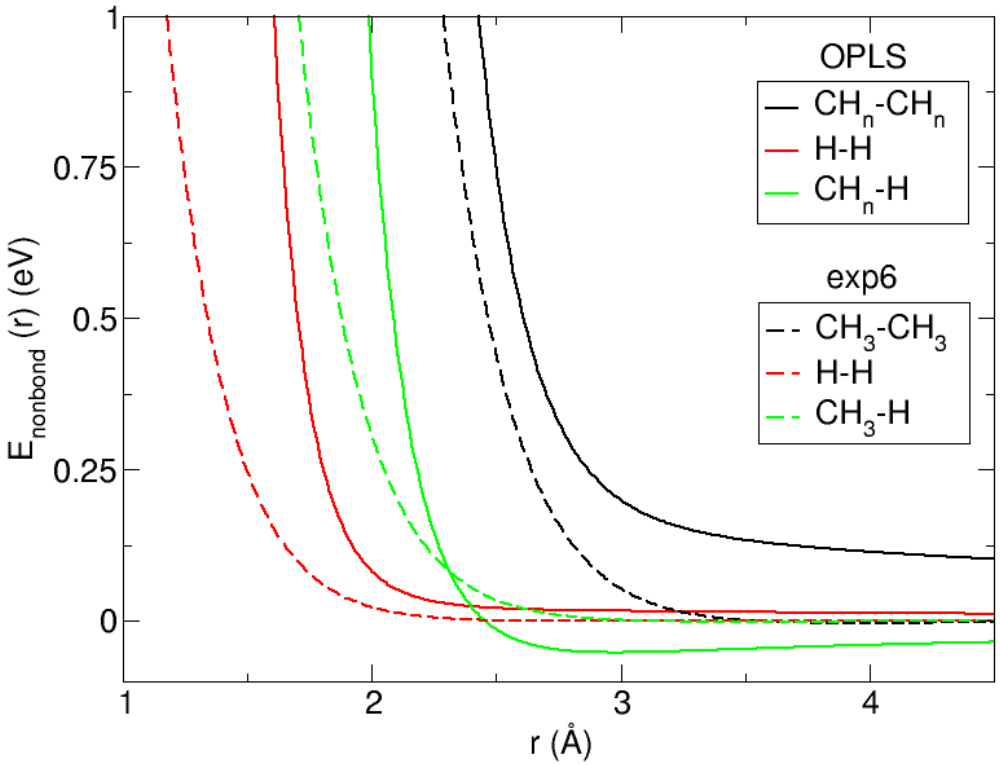
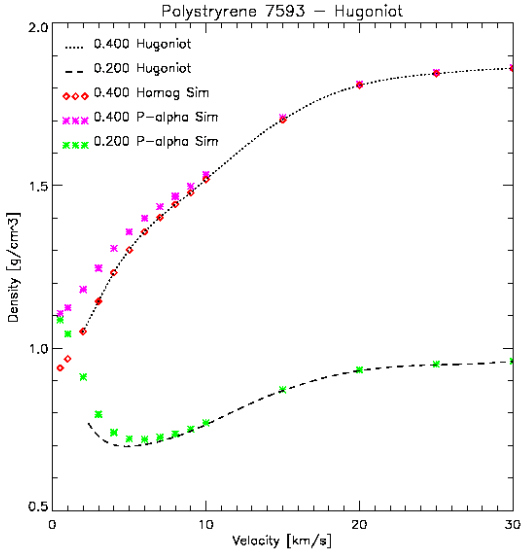
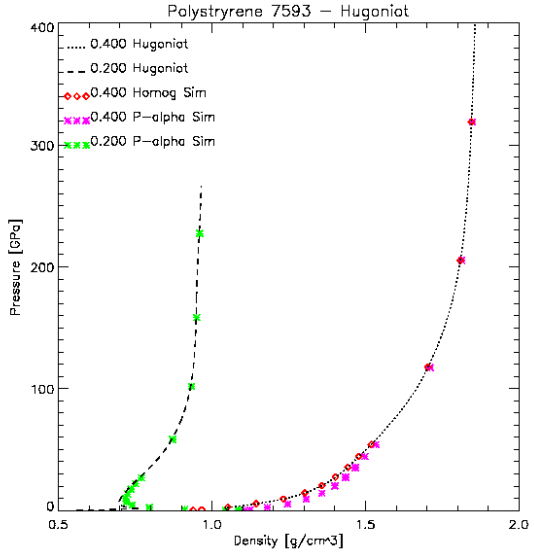
OPLS and Borodin-Smith utilize different repulsive functional forms

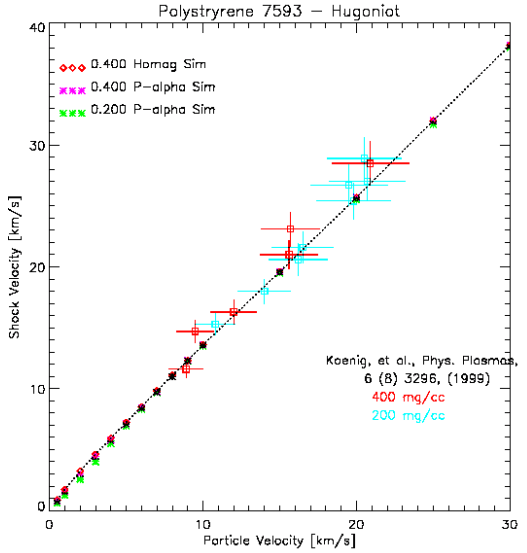


- AIREBO and OPLS share L-J 12-6 functional form for repulsive interaction
- Borodin-Smith has a weaker exponential form, and a more physical shock behavior
- Shock-problems probe regions of the potential far from the equilibrium region -- where they are parameterized and exhibit very similar characteristics
- *Behavior under shocks are difficult to predict using potentials mainly for normal state*
- *DFT-QMD/AM05 yields results of high fidelity for shocked polymers -- non-trivial finding*

Validation of P-α Model for Polystyrene (2 of 2)



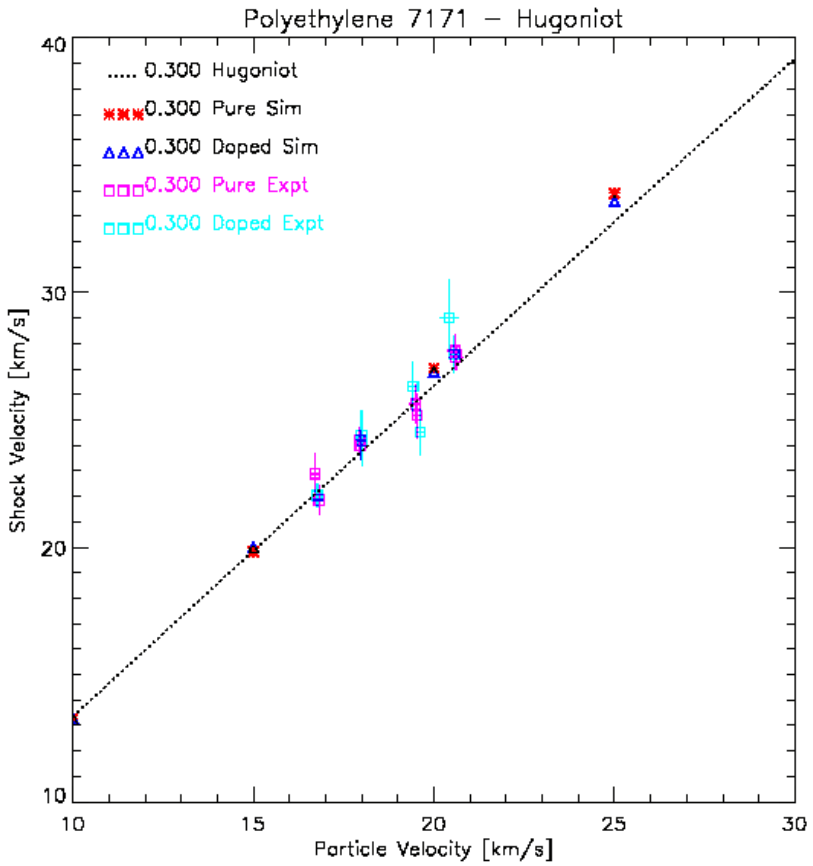




- **Largest difference between P-α and homogenous foam models seen in density**
- **P-a compresses to higher density**
- **Aneos 7593 (and 7593) over compresses 200 mg/cc foam at low velocities**
  - **Reason unknown at present**

- **Us-Up relation in good agreement between experiment and simulation**

Homogenous pure and platinum-doped foam simulations shock to same pressures and agree with Z experiments



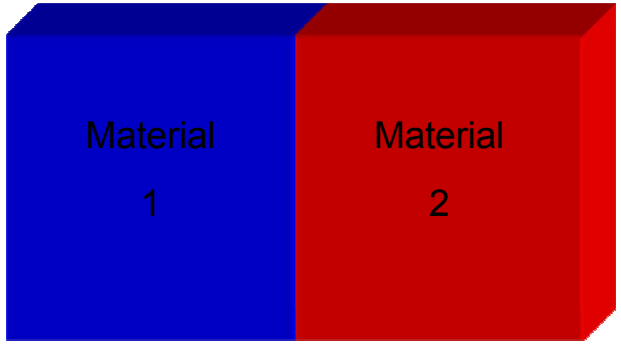
- Undoped and doped foams nominally 0.300 g/cm^3 average density
- Platinum-doped foam is 50-50 mixture by weight
  - 99.3% foam and 0.7% platinum by volume
- Simulations use P-α model for foams
- Doped foams shock to same pressures at undoped foams
  - Uncertain behavior at low flyer velocities (< 7 km/s) for doped foams

EOS Mixing Rules (aka Multi-Material Treatment) in ALEGRA (part 1 of 3)

- Applies when two (or more) materials are present within a single computational mesh element (cell)
- Normal values for all materials
  - Volume fractions, f_1 and f_2
  - Densities, ρ_1 and ρ_2
  - Temperatures, T_1 and T_2
  - Pressure, $P_1(\rho_1,T_1)$ and $P_2(\rho_2,T_2)$
  - Specific Energy, $e_1(\rho_1,T_1)$ and $e_2(\rho_2,T_2)$
- Initial Conditions
  - e and P from ρ and T
- Simulation Update
  - T from ρ and e
  - P from ρ and T
- *No explicit material interface*
  - *Inferred or reconstructed for remap*

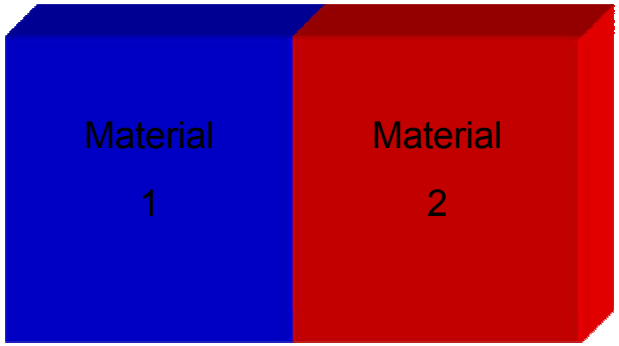
Mixing rules:

- $1 = f_1 + f_2$
- $\rho = f_1 \rho_1 + f_2 \rho_2$
- $T = (f_1 \rho_1 C_{v1} T_1 + f_2 \rho_2 C_{v2} T_2) / (f_1 \rho_1 C_{v1} + f_2 \rho_2 C_{v2})$
- $P = f_1 P_1 + f_2 P_2$
- $e = f_1 \rho_1 e_1 + f_2 \rho_2 e_2$

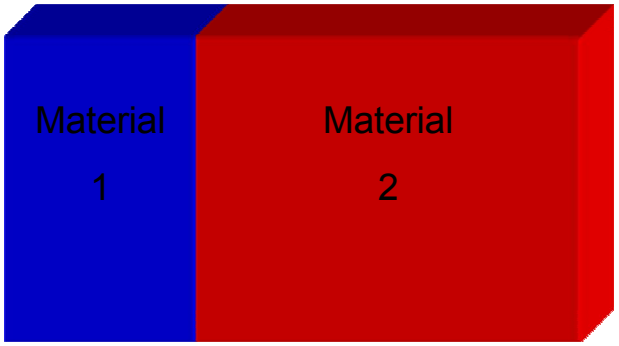


EOS Mixing Rules (aka Multi-Material Treatment) in ALEGRA (part 3 of 3)

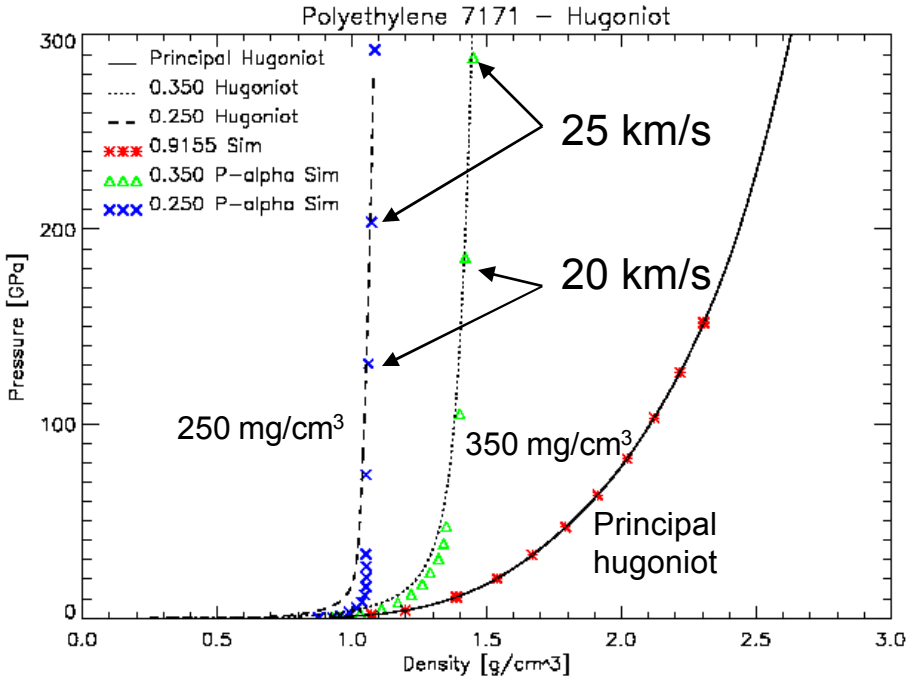
- Default (legacy) method:
  - $P = f_1 P_1 + f_2 P_2$
  - No equilibration of pressure
    - » Pressure differences not taken into consideration
  - No relaxation of temperature differences
    - » (Thermal conduction performs temperature equilibration)
  - Easily leads to unphysical results
- Constant volume fractions still assumed in expansion

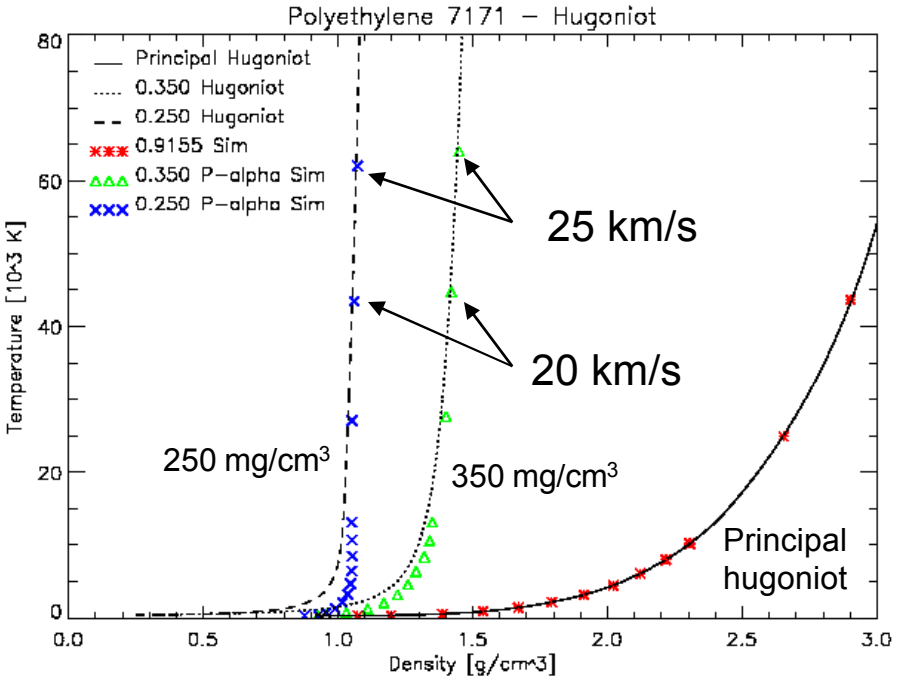


- Isentropic Compression Method:
  - $P = B\,(f_1 P_1 + f_2 P_2)$
  - $B = (f_1/B_1 + f_2/B_2)^{-1}$
  - An energy equation also solved to account for compression or relaxation
  - Soft materials compress more
  - Stiff materials compress less
  - Optional thermal relaxation available
  - *Additional details for robustness*



Piston driven foam with virtual flyer – Shocked foams expected to reach Mbar pressures

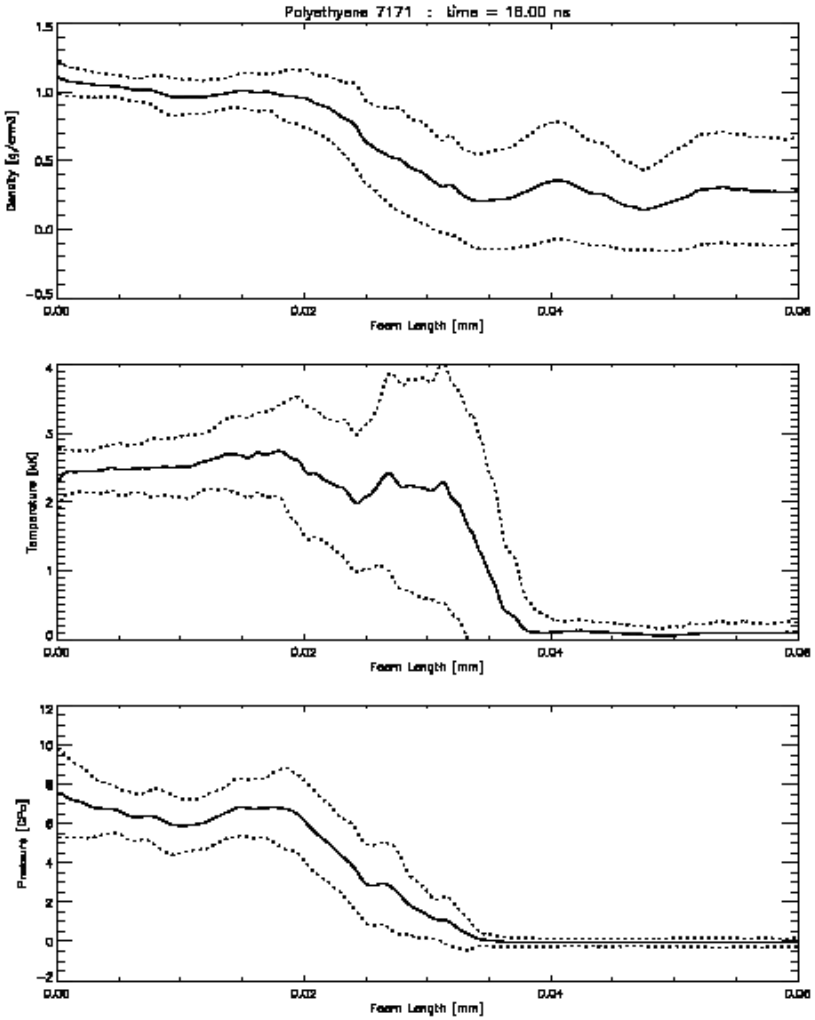




- **Hugoniot simulations use P-alpha model for polyethylene foam**
  - **P-alpha model compresses foam to near solid density before rising significantly**
  - **Asymptotes to homogeneous foam hugoniot at high pressures and temperatures**

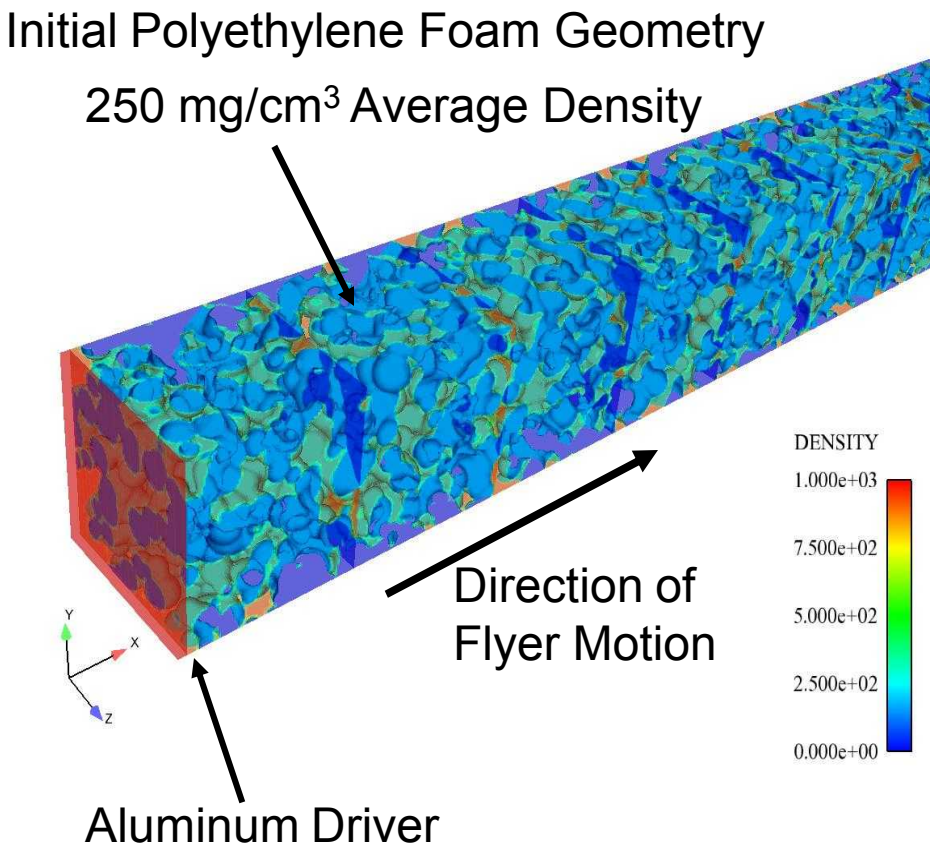
Piston driven foam with virtual flyer – Simulated as a Noh problem

- **Change frame of reference to that of the piston**
  - **Rigid wall (piston face) remains stationary**
  - **Foam runs into rigid wall**
  - **Piston/foam interface otherwise unstable**



Piston driven foam with explicit flyer

- **Initial simulations use a constant velocity aluminum piston as a driver**
  - **Will replace with real MHD driven aluminum flyer later**
- **Solid density polyethylene block is permeated with very low density polyethylene spheres (voids) to obtain desired average density**
  - **1 to 2 micron pore size**
- **Mesh and material is doubly periodic in y & z**
  - **20 x 20 micron domain in y & z**
  - **200 micron foam length in x**
  - **Vary 0.05 to 0.25 micron cell size to check mesh resolution**
- **Will compare to experimental gas gun and Z data**
  - **Will validate the P-alpha models**



Piston driven foam with explicit flyer

- Test simulations at 5 km/s
- Piston/foam interface is unstable
- Present thinking is that the phenomenon is real
  - Word is that VISAR records are “incoherent” for the foam experiments on Z at 20 to 25 km/s
  - Even at 5 km/s pressures in the aluminum flyer exceed the yield strength
  - Aluminum is therefore very plastic
  - Enhancing aluminum strength by factor of 100 mitigates interface instability