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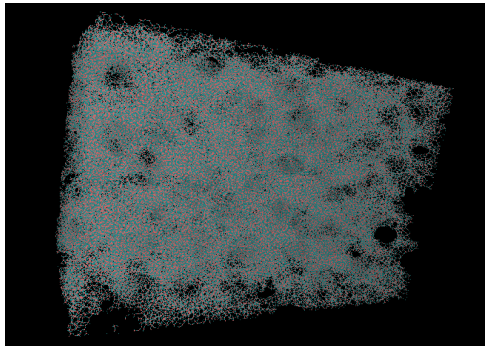

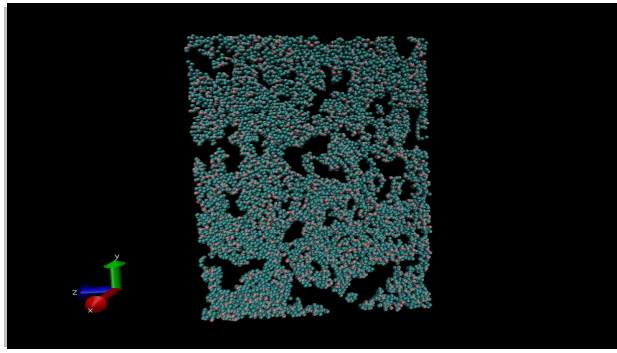

# Molecular dynamics study of shock compression in porous silica glass

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U.S. DEPARTMENT OF ENERGY

NNSA National Nuclear Security Administration

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# Overview

- Motivation
- Methodology
- Results
- Conclusions

# Motivation for studying porous silica

- Low density silica aerogels with a range of initial densities are commonly used in extreme conditions:
  - Space missions [1, 2]
  - High P research [3, 4, 5]
- Gaining a more detailed understanding of the processes occurring under these conditions is important.
- Validate model by comparison with experiment.

Experimental data

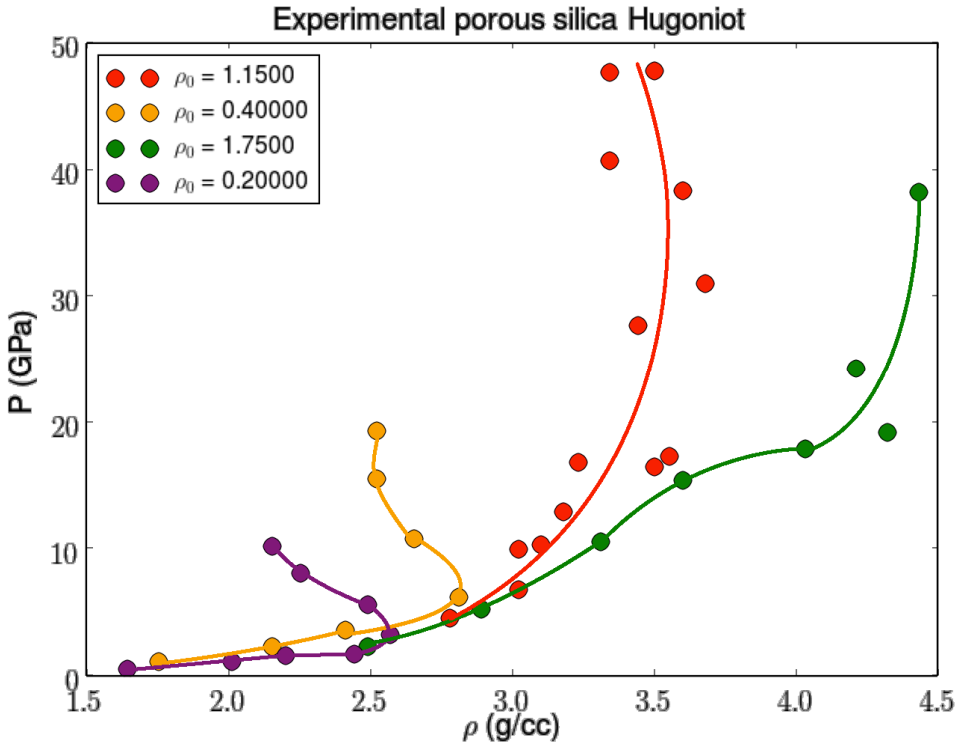

1. M. Burchell et al., *Annu. Rev. Earth Planet. Sci.* **34**, 385 (2006).
2. D. Brownlee et al., *Science* **314**, 1711 (2006).
3. M. D. Knudson, J. R. Asay, and C. Deeney, *J. Appl. Phys.* **97**, 073514 (2005).
4. N. Holmes, High-Pressure Science and Technology—1993 (AIP, New York, 1994), p. 153.
5. M. D. Knudson and R. W. Lemke, *J. Appl. Phys.* **114**, 053510 (2013).

# Motivation for studying porous silica

An anomalous density inversion in the Hugoniot is observed experimentally at high shock pressures for highly porous silica [6]. This is due to shock energy being converted into kinetic energy as particles are vaporized into the void space. Thermal expansion occurs with respect to the same system shocked to a lower pressure. Local heating during void collapse has been studied with molecular dynamics (MD) [7].

Can we model this density inversion with molecular dynamics?

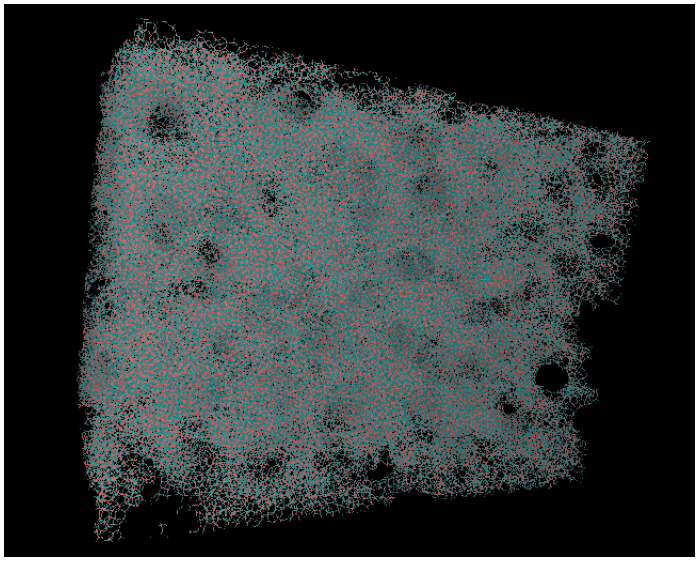
Can we model this density inversion without using the expensive, propagating, non-equilibrium molecular dynamics (NEMD) model for dynamical shock simulation?




Density inversion in highly porous silica Hugoniot is not well studied with MD.


6. R. F. Trunin, *Experimental data on shock compression and adiabatic expansion of condensed matter* (2001).
7. J. Matthew D. Lane et al., *Comp. Mat. Sci.* ***79****, 873-876 (2013).*

# Methodology - General



| $\rho_0$ (g/cc) | % Porosity |
|---|---|
| 2.212 | 0 |
| 2.09 | ~5 |
| 1.08 | ~50 |
| 0.56 | ~75 |
| 0.23 | ~90 |
| 0.14 | ~95 |

A range of silica systems with various porosities (initial densities) have been created in LAMMPS [8], molecular dynamics package.

Molecular dynamics at the fully atomistic level is employed. Each atom is treated as a partially charged point particle moving under the influence of a user specified potential energy surface:

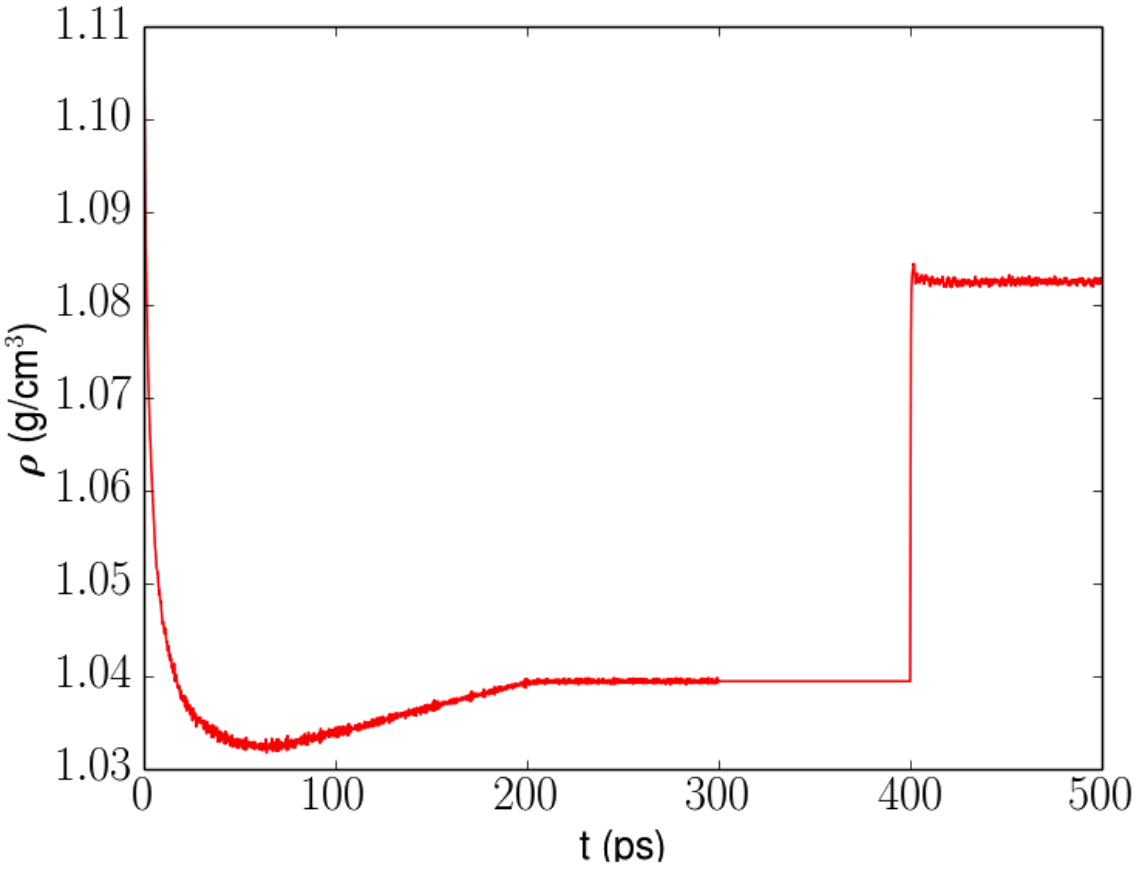
Short/medium range interactions are calculated using the $SiO_2$ BKS potential [9], a simple, tabulated interatomic potential for silica.
Long range interactions are calculated using an Ewald summation.

Each porous silica system is shocked to a range of pressures from the reference state in order to build the Hugoniot.


8. S. Plimpton, *J. Comp. Phys.* **117**, 1-19 (1995).
9. B. W. H. van Beest et al., *Phys. Rev. Lett.* **64**(16), 1955 (1990).

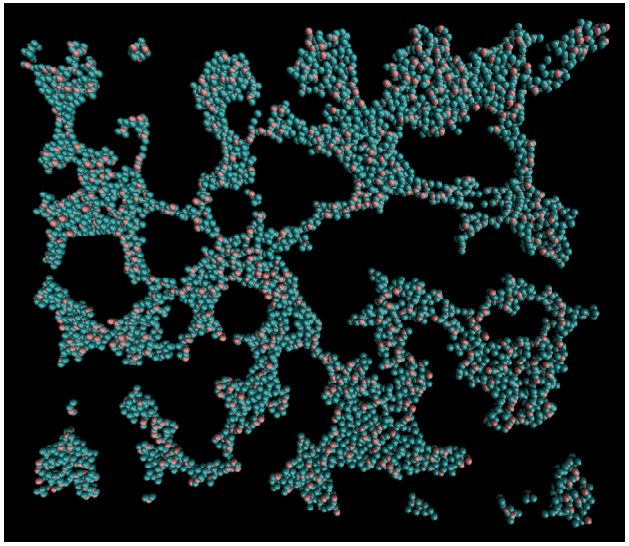
# Methodology – Initial System

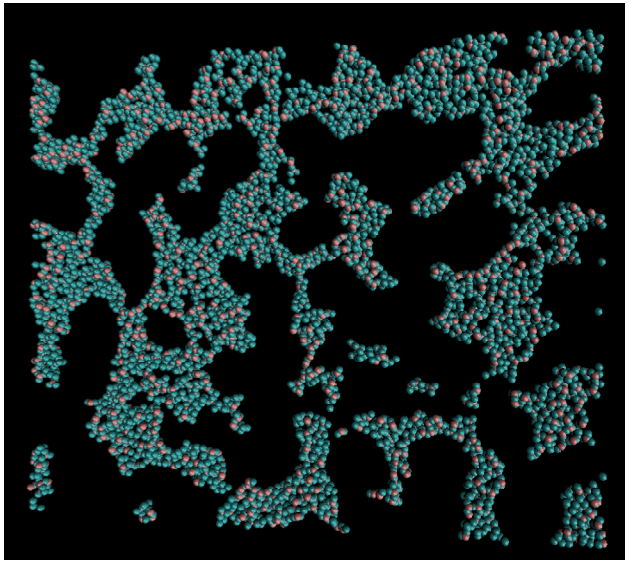
- Expand spherical voids at 2500 K until desired porosity is reached
- Cool to 300 K.
- Equilibrate
- Remove voids and re-equilibrate.

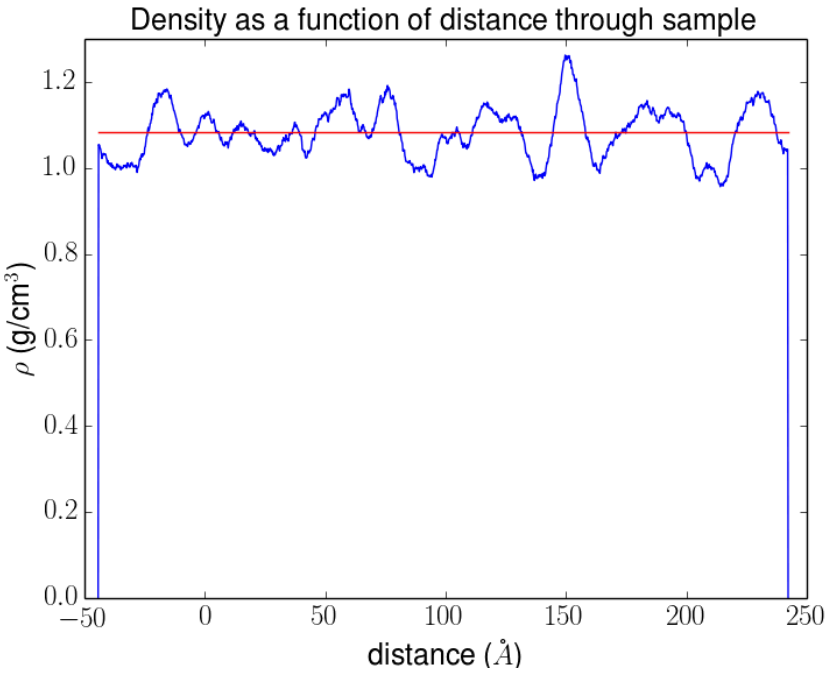



~50 % porous system
Periodic boundary conditions
Timestep: 1 fs
Average density: 1.08 g/cc
Average pore diameter: 20.0 Å
Number of atoms: 373248
Box dimensions: 182 x 217 x 290 $Å^3$

# Methodology – Initial System



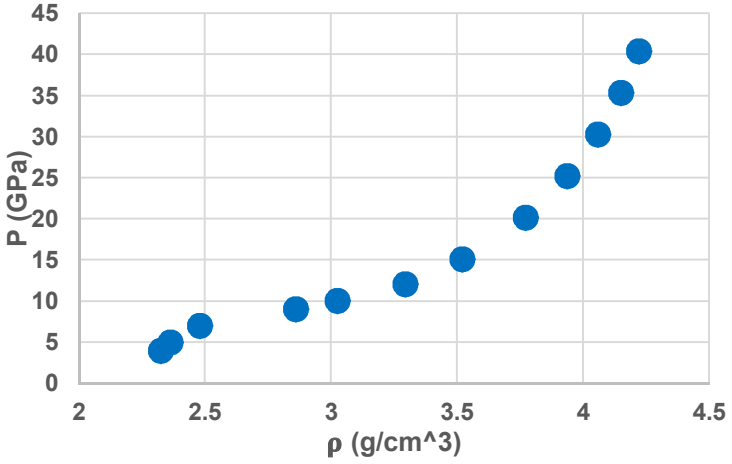





Each data point - 4 Å slice throughout 50 % porous sample
Average density: 1.08 g/cc
Average pore diameter: 20.0 Å
Number of atoms: 373248
Box dimensions: 182 x 217 x 290 $Å^3$

# Methodology – Reproducing the Hugoniot in MD

Hugoniot: the locus of points representing a series of final states originating from a single reference state, when the final and reference states are separated by a discontinuity (a shock), satisfying the jump conditions (energy, mass, and momentum conservations) which relate the initial state to the final state.




Reproducing the final state in MD

1. Non-equilibrium molecular dynamics. Propagate a shock through the material via a momentum mirror [7,10], calculate state variables to verify jump conditions.
   - Expensive and requires large system sizes.
   - Effectively reproduces experimental Hugoniot data for porous systems [7].

2. Non-propagating, constant stress Hugoniostat method [11]. Uniaxially compress system until final pressure is reached and jump conditions are met.
   - Less expensive.
   - Well tested in traditional Hugoniot space.
   - Not guaranteed to evolve along Rayleigh line or any other path through ρ, P space.
   - Path and efficiency of path depend on damping coefficients.
   - Previously untested on porous systems.


10. B. L. Holian and P. S. Lomdahl, *Science* **280**, 2085 (1998).
11. R. Ravelo et al., *Phys. Rev. B* **70**, 014103 (2004).

# Methodology – Reproducing the Hugoniot in MD.

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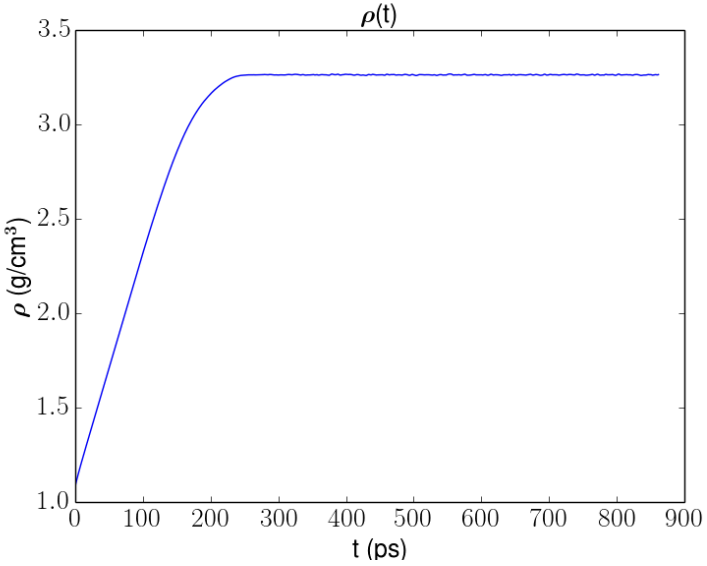
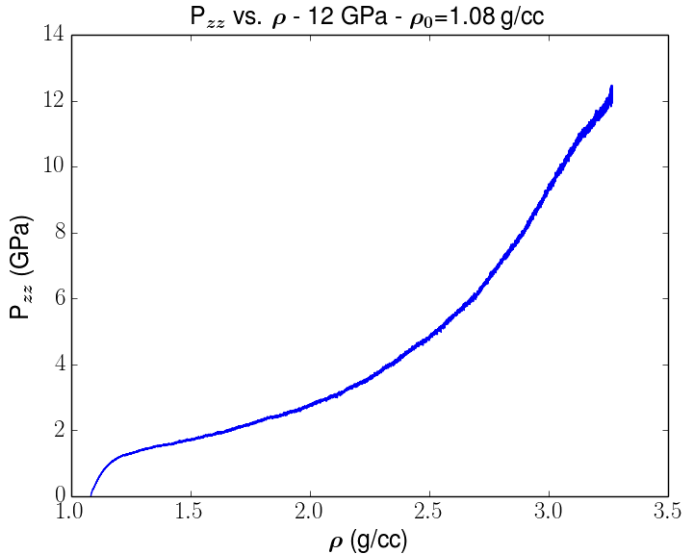
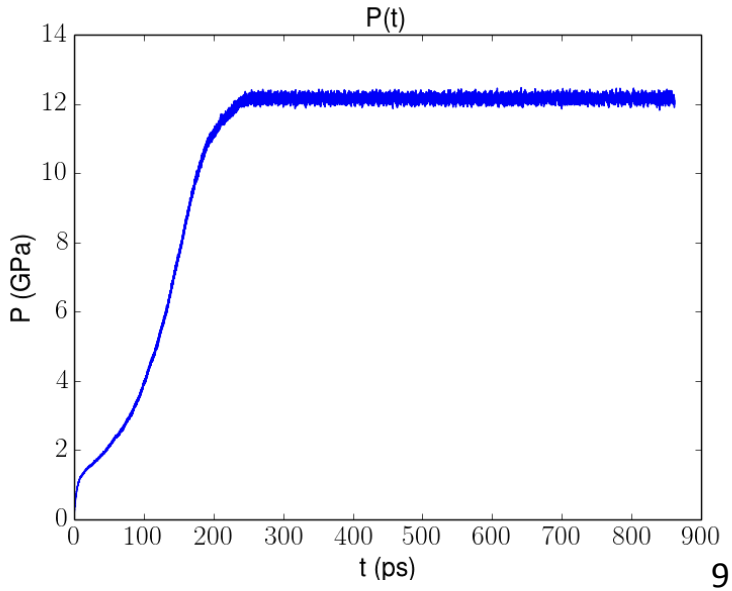
Constant stress Hugoniostat method in action.
50 % porous (1.08 g/cc) silica.
Final pressure: 12 Gpa.
Time step: 0.2 fs.
Temperature damping coeff: 20 ps
Pressure damping coeff: 20 ps

<u>Trajectory:</u>



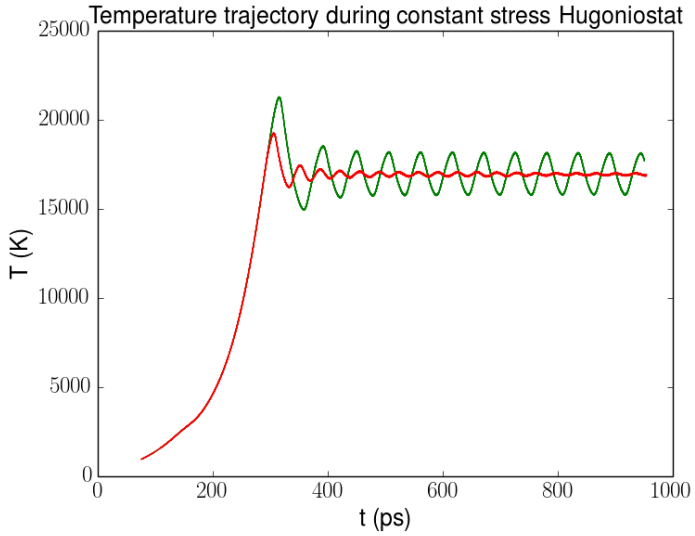



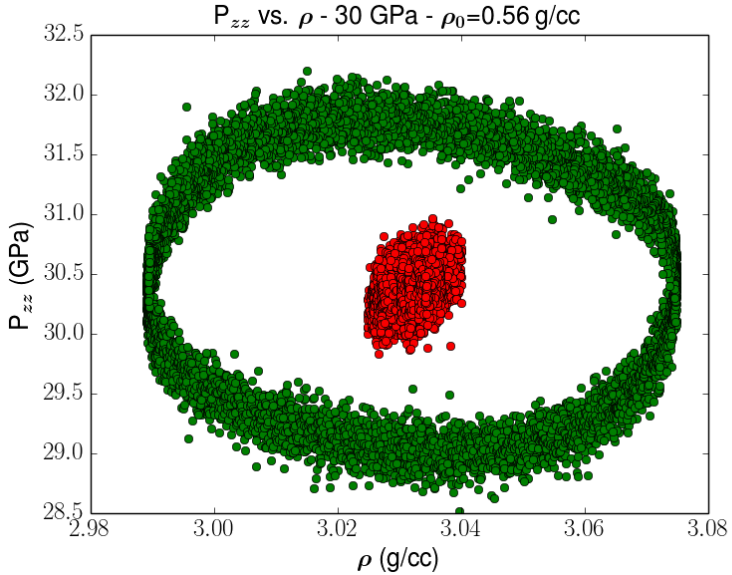



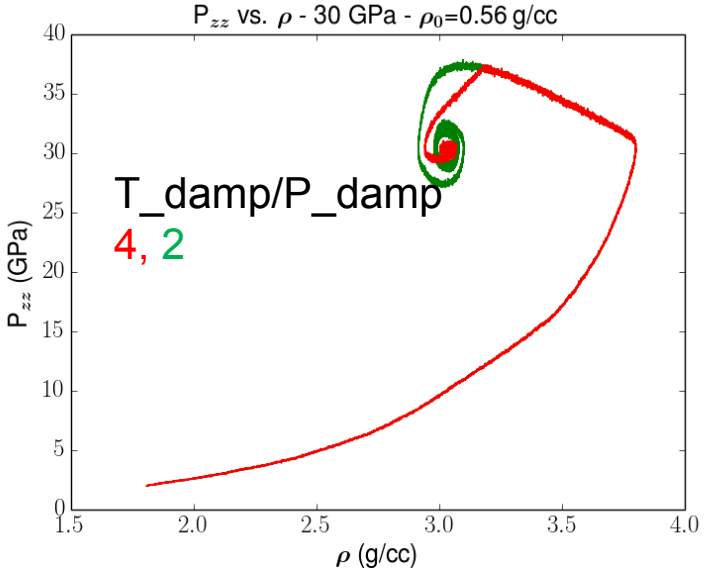
# Methodology – Reproducing the Hugoniot in MD.

75 % porous (0.56 g/cc) silica.
Final pressure: 30 Gpa.
Time step: 0.2 fs.

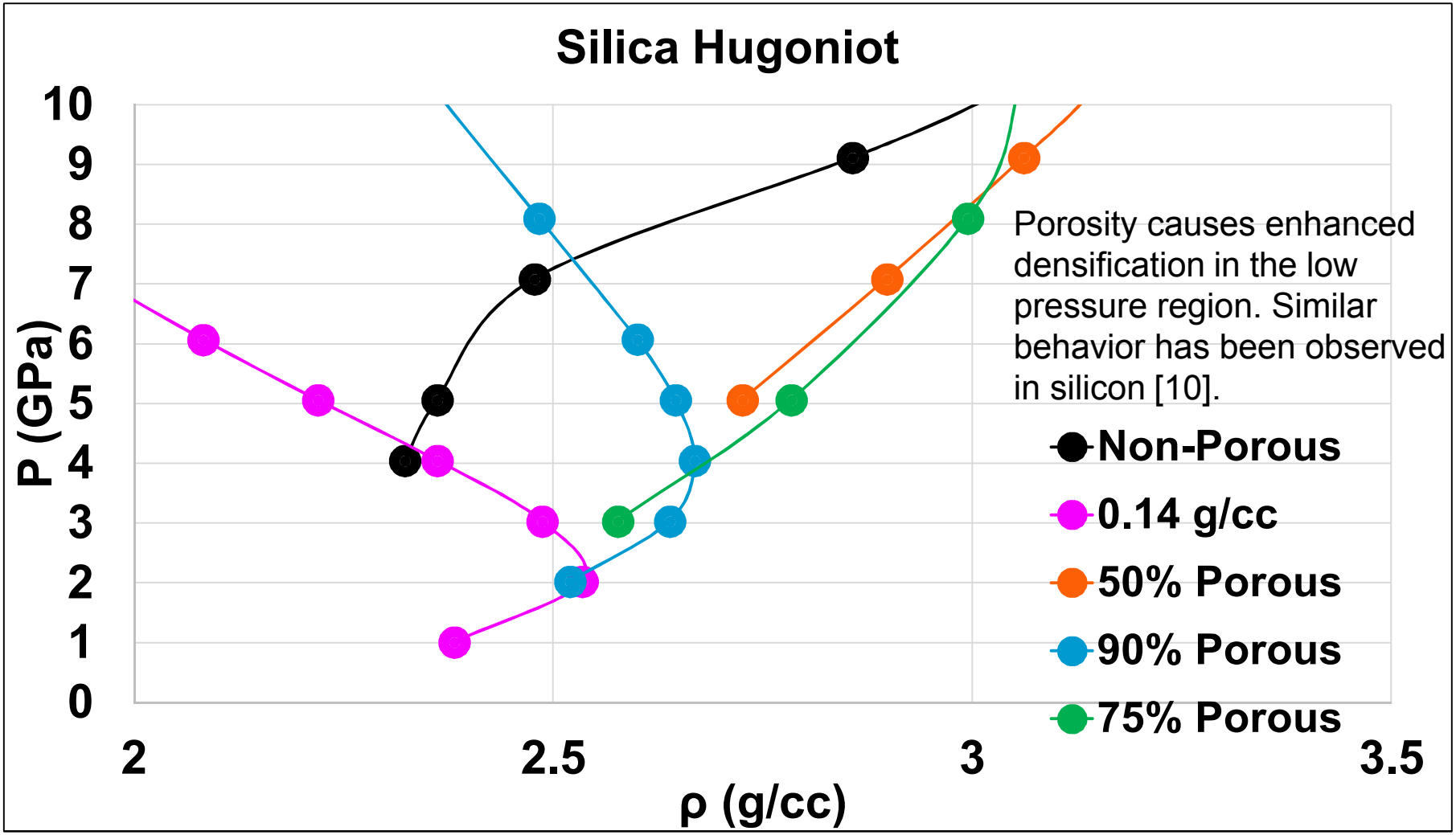
System gets very hot (~15000 K) compared to an analogous final pressure for fully dense silica (~2000 K). Final state is not very sensitive to damping coefficient ratios.



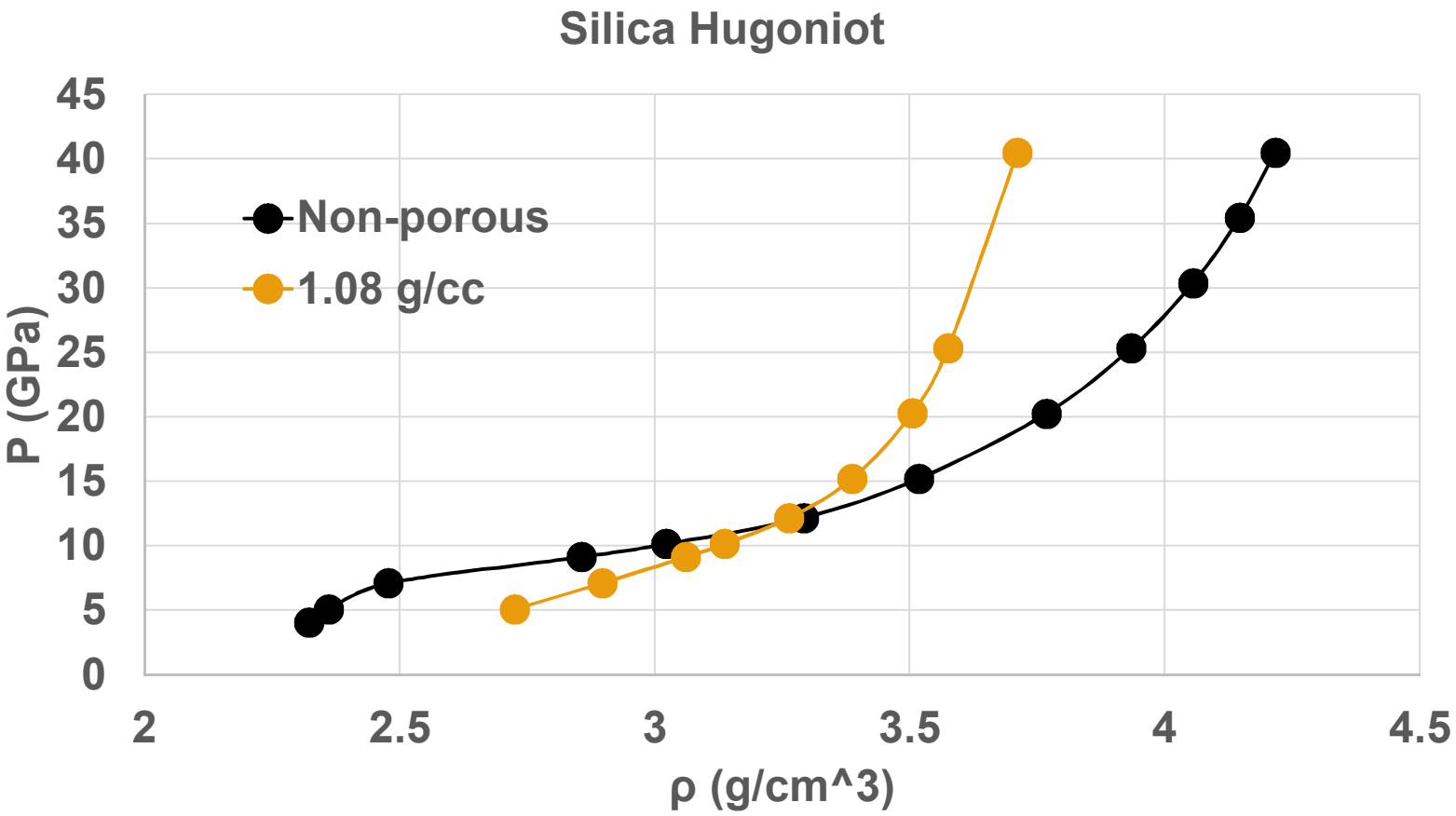



# Results – Enhanced densification



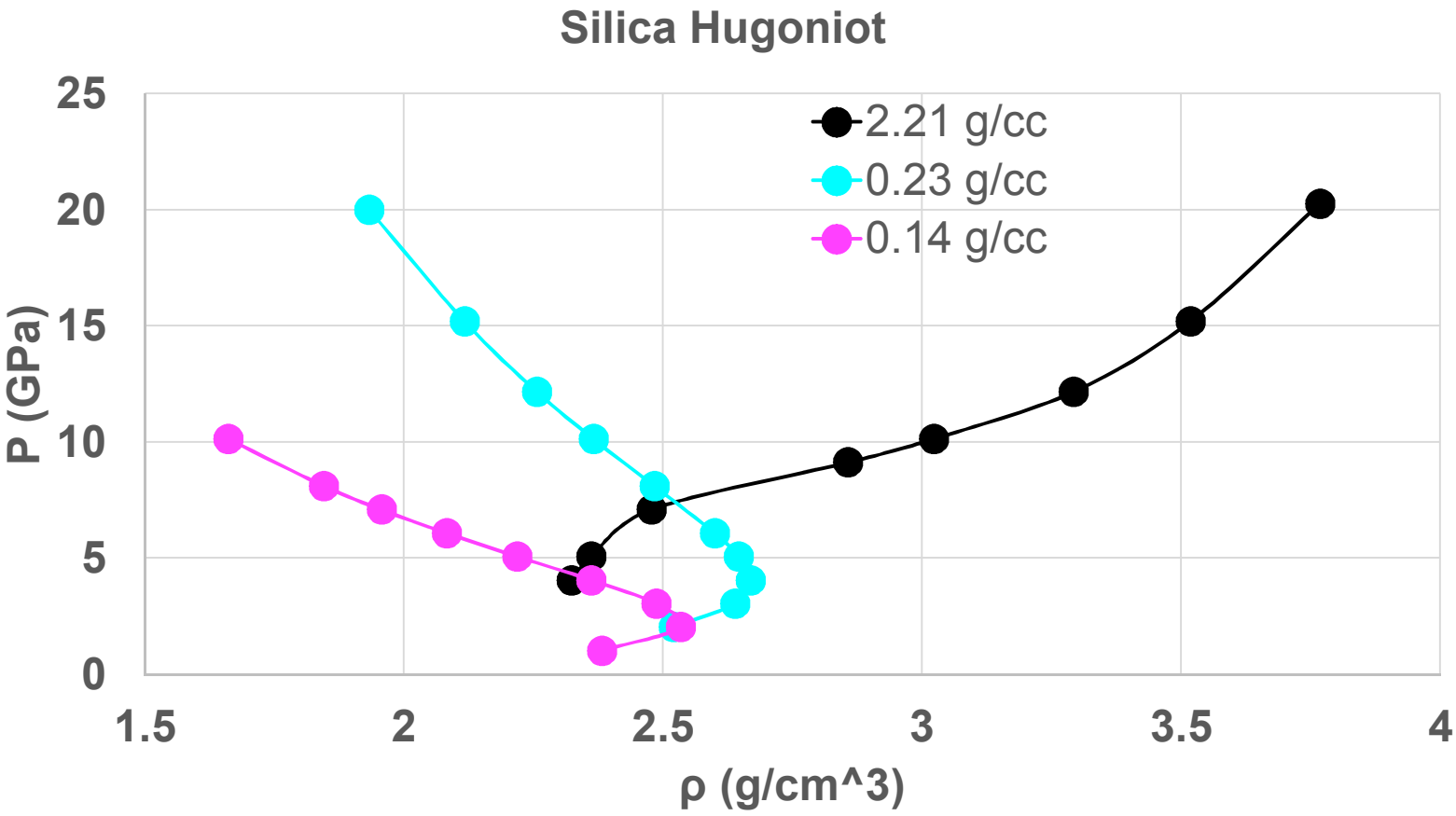

10. J. Matthew D. Lane et al., *Phys. Rev. B* **90**, 134311 (2014)

# Results – Enhanced densification



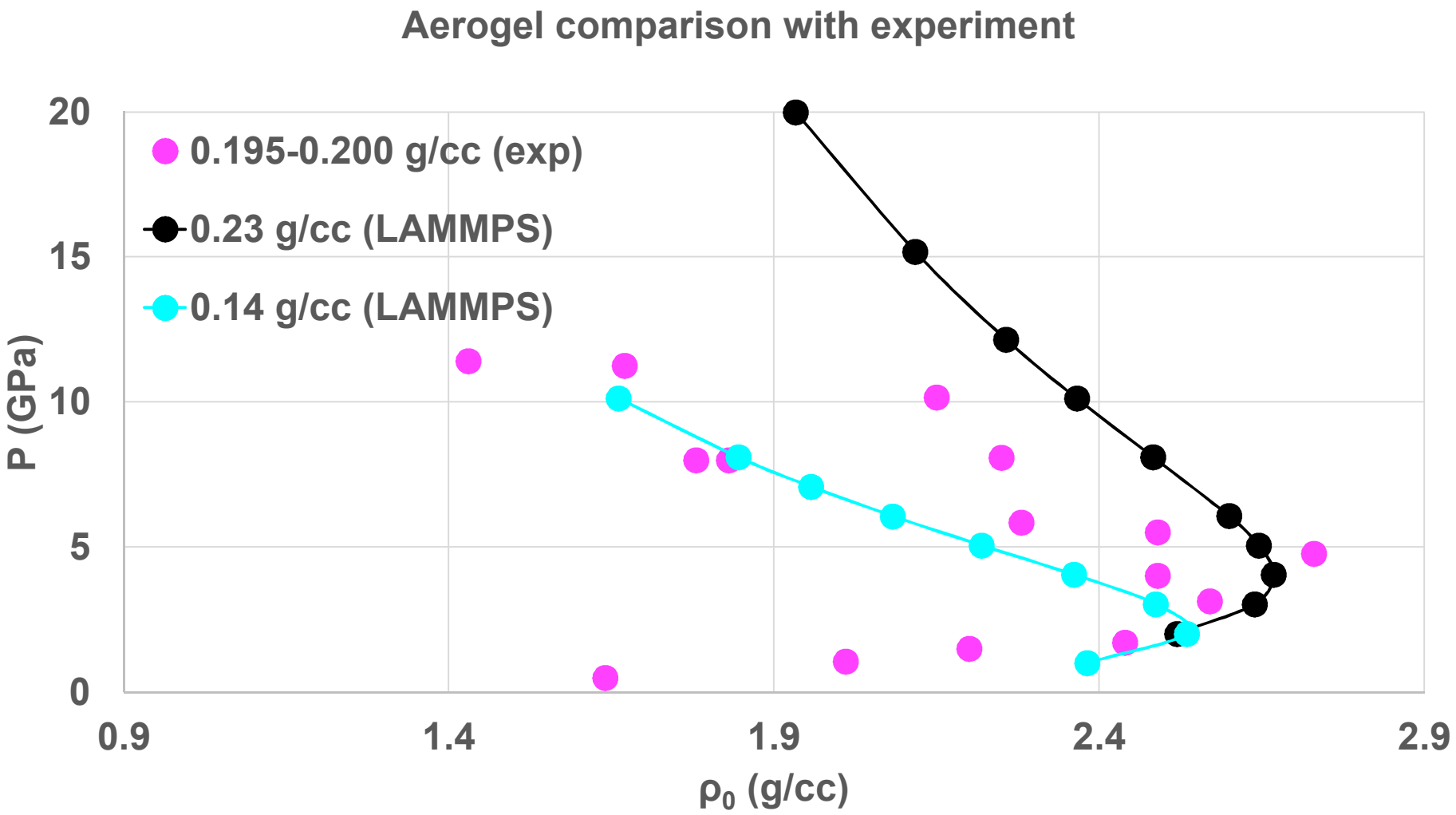

Enhanced densification is observed at lower pressures.
Normal behavior of shocked porous material relative to shocked non-porous material returns above 10 Gpa.

# Results – Density inversion

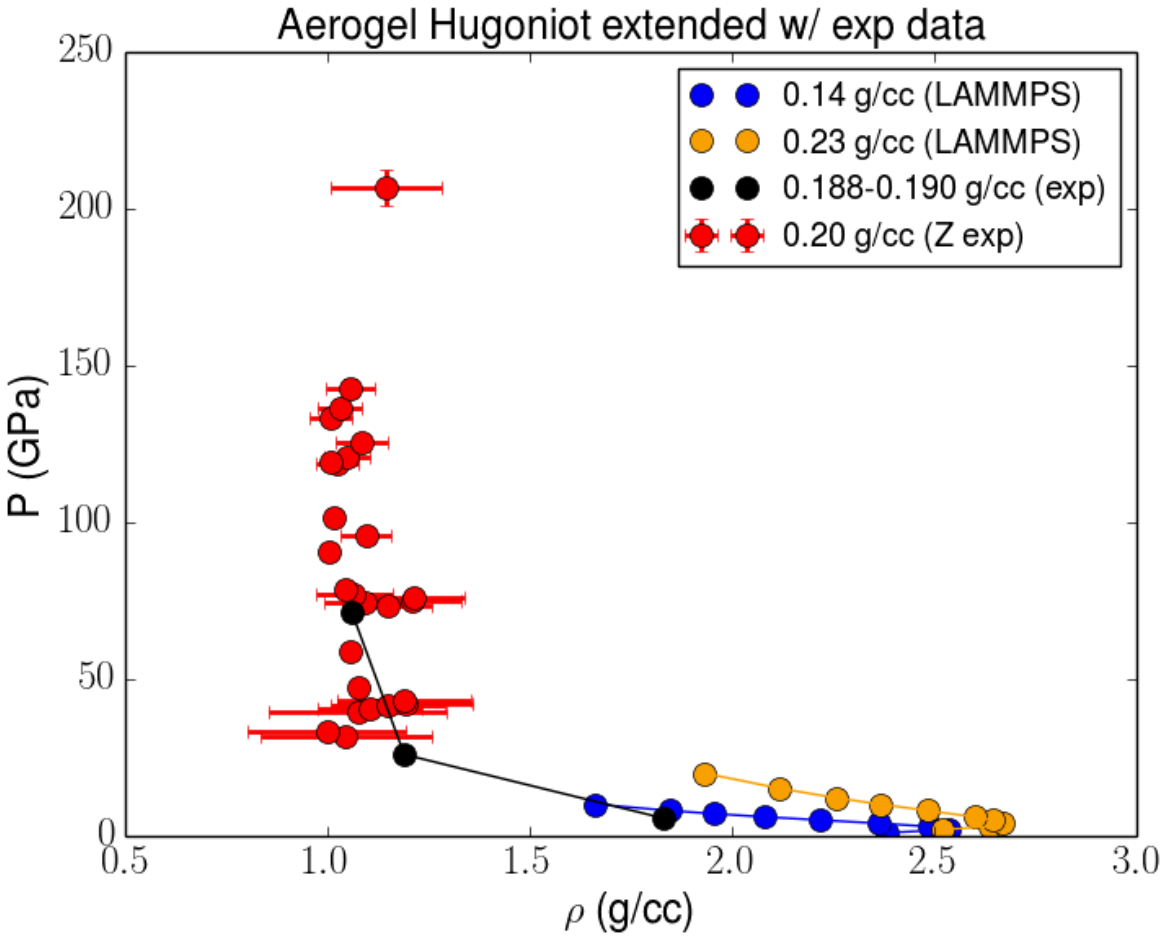



Demonstrated density inversion is in qualitative agreement with experiments

# Results – Density inversion



# Results – Density inversion




Very high pressure experimental data for highly porous silica, collected using the Z-machine [5], plotted against LAMMPS and other experimental data shows a cusp in the Hugoniot.

Z machine data is not reproducible with molecular dynamics. Simulation at the level of density functional theory is needed.

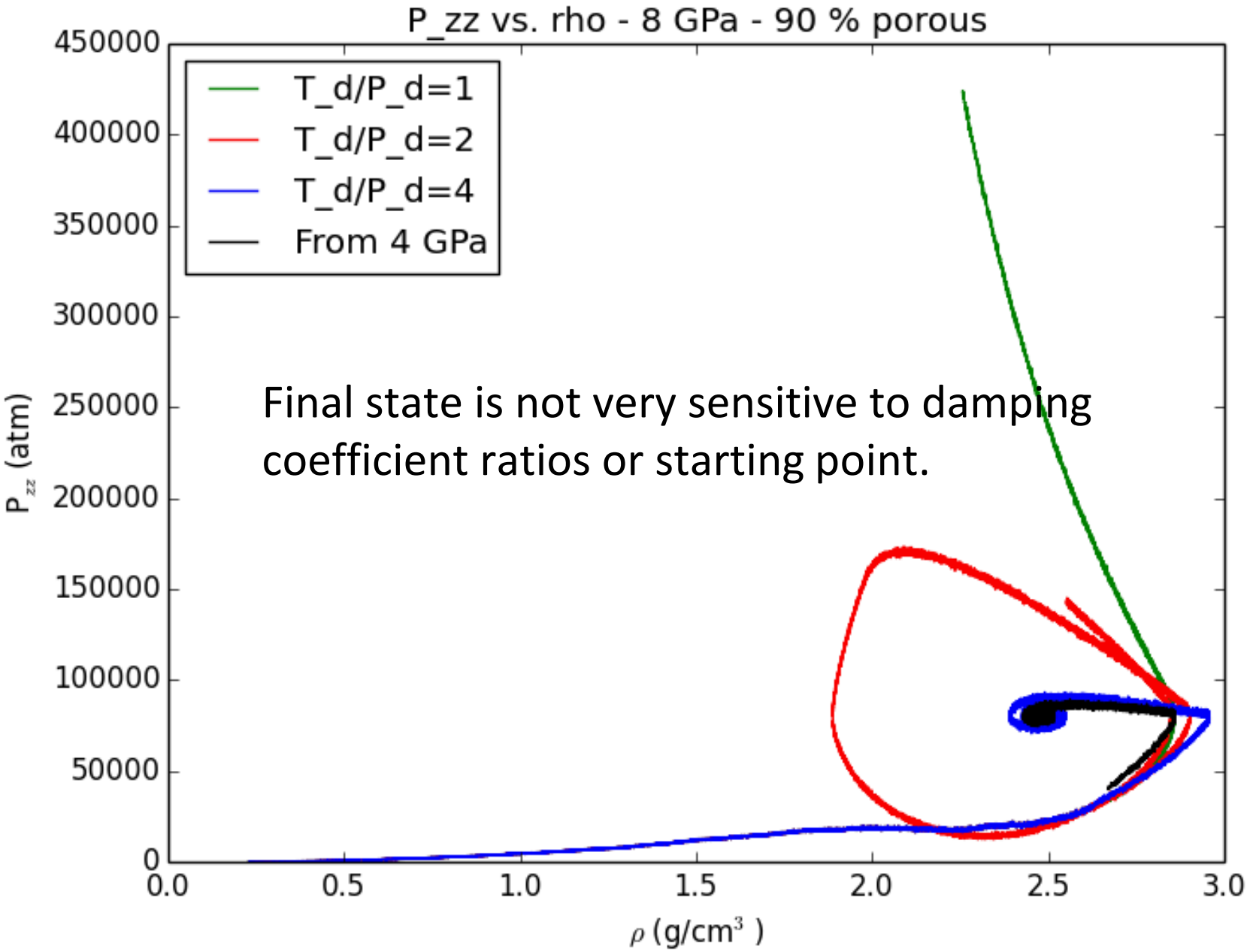

5. M. D. Knudson and R. W. Lemke, *J. Appl. Phys.* **114**, 053510 (2013).
6. R. F. Trunin, *Experimental data on shock compression and adiabatic expansion of condensed matter* (2001).

# Conclusions

- BKS, a very simple $SiO_2$ potential, coupled with the constrant stress Hugoniostat reliably reproduces the Hugoniot for porous $SiO_2$.
- Enhanced densification is observed at lower pressures.
- Negative $dP/d\rho$ in the Hugoniot for highly porous $SiO_2$ is qualitatively captured in this very simple model.

# Acknowledgements


- Sandia National Labs
  - High Performance Computers
- Kyle Cochrane
- Thomas Mattsson



P_zz vs. rho - 8 GPa - 90 % porous
T_d/P_d=1
T_d/P_d=2
T_d/P_d=4
From 4 GPa
Final state is not very sensitive to damping coefficient ratios or starting point.
P_zz (atm)
ρ (g/cm³ )
450000
400000
350000
300000
250000
200000
150000
100000
50000
0
0.0
0.5
1.0
1.5
2.0
2.5
3.0

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# 50 % void radii distribution In angstroms

| Average radius | | 9.96225607 |
|---|---|---|
| Range | N voids | |
| 4 to 5 | 23 | |
| 5 to 6 | 333 | |
| 6 to 7 | 290 | |
| 7 to 8 | 292 | |
| 8 to 9 | 320 | |
| 9 to 10 | 277 | |
| 10 to 11 | 281 | |
| 11 to 12 | 291 | |
| 12 to 13 | 311 | |
| 13 to 14 | 288 | |
| 14 to 15 | 285 | |
| 15 to 16 | 38 | |