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Hugoniot Simulations of Porous Tantala (Ta_2O_5)

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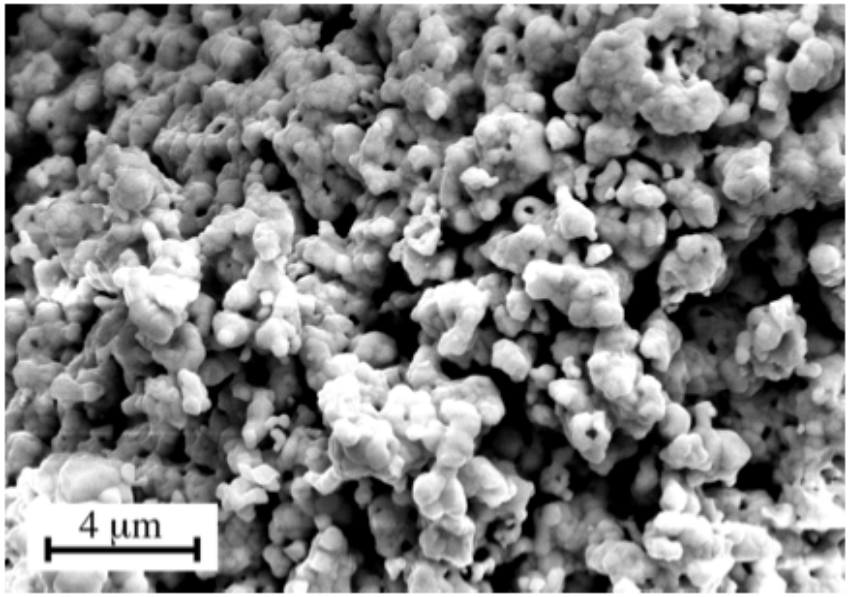
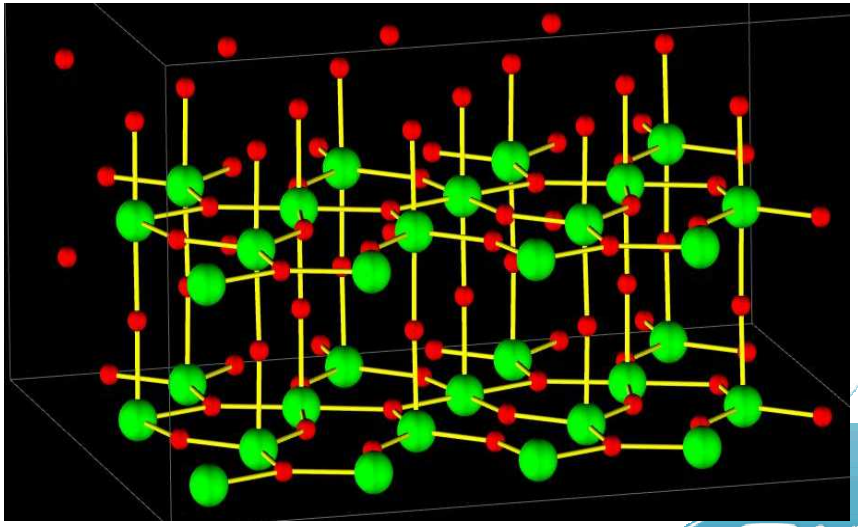
Porous materials are everywhere

- **Many types of porous materials**
  - Rocks/Soil
  - Cork
  - Foams
  - Ceramics
- **Uses**
  - Petroleum Engineering
  - Civil Engineering
  - Electronics
  - Filters
  - Padding/Cushions
- **Even a Journal "Advanced Porous Materials"**
- **Using Shocks on these porous materials allow us to explore a larger domain of phase space.**

Porous Tantala





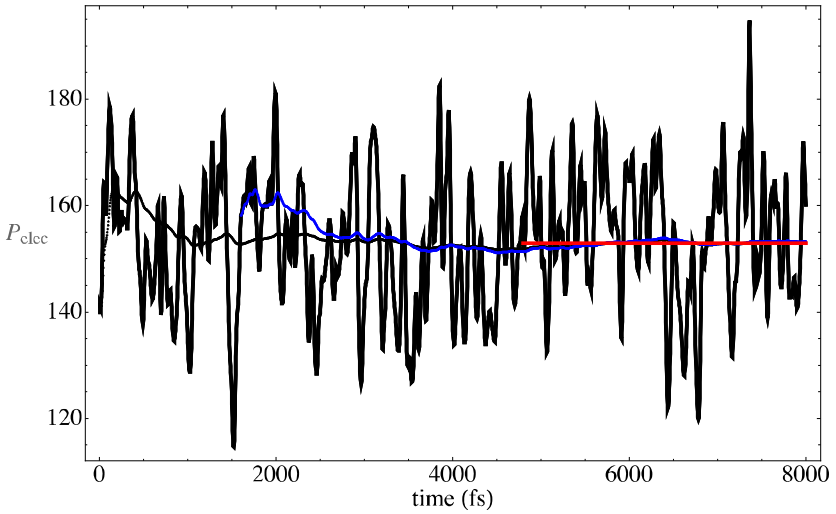
R. Ramprasad, J App Phys, Vol. 94, No. 9. (Nov 2003)

Using DFT/QMD to model porous tantala

- **Computational setup**
  - –VASP
  - –Basis cell
  - –Simulation methods
- **Gruneisen Gamma simulations**
  - –Gruneisen EOS
- **Hugoniot Calculations**
- **Correcting for surface energy**
- **Comparison with Data**
- **Conclusions**

Assemble a reference system and use molecular dynamics to equilibrate.

- **First-principles simulations DFT**
  - VASP – plane-wave code w PAW core-functions
  - Use of DFT codes simulating warm dense matter
    - *M. P. Desjarlais Phys. Rev. B **68**, 064204(2003)*
  - Great care in convergence
    - *A. E. Mattsson et. al. Modelling and Simulation in Material Science and Engineering **13**, R1 (2005)*
- **Assemble reference system**
  - 32 tantalum atoms and 80 oxygen atoms.
  - Baldereschi mean value k-point (¼, ¼, ¼)
  - LDA potential with 11 electron tantalum pseudo potential and 6 electron oxygen potential
  - Allowed to equilibrate to a constant mean pressure and energy
  - Equilibrated for multiple ps
  - Cutoff energy at 600 eV
  - Standard deviation of energy and pressure <1%
  - Block averaging to reduce correlation



Quantum molecular dynamics (QMD) simulations give thermo-physical properties

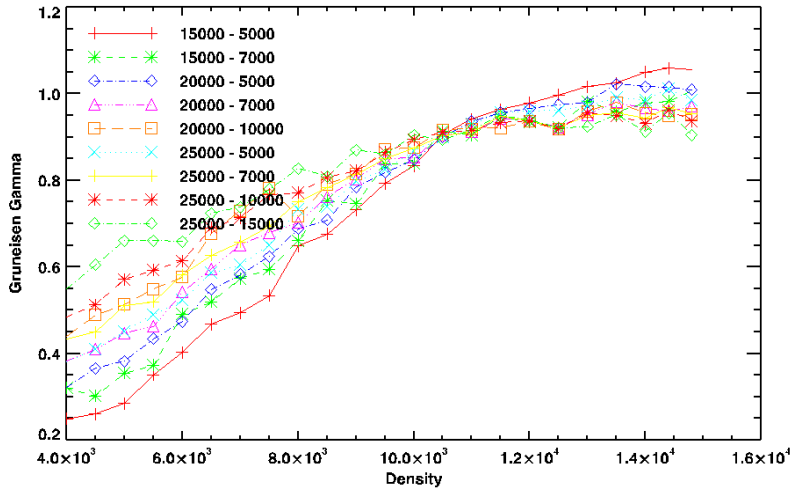
Calculate a broad range of pressures and energies for Gruneisen Γ

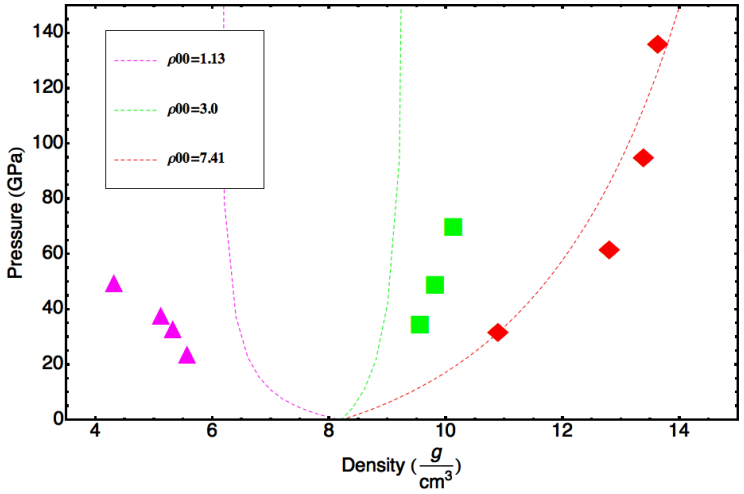
- **A broad range of pressure and energy isotherms**
- **Calculate Γ from** $\Gamma = V \dfrac{dP/dT}{dE/dT}$
- **Γ is not constant until slightly compressed**
- **Mie-Gruneisen EoS**

$$P_H = \frac{\rho_o C_o^2 \chi \left[1 - \frac{\Gamma}{2}\chi\right]}{(1-\eta\chi)^2} + \Gamma \rho E \qquad \chi = 1 - \frac{\rho_o}{\rho} \qquad \eta = dU_s / dU_p$$

$$P_{H,P} = \frac{P_H\left(1 + \frac{\Gamma}{2}\left(1 - \frac{\rho}{\rho_0}\right)\right)}{1 + \frac{\Gamma}{2}\left(1 - \frac{\rho}{\rho_{00}}\right)}$$

- **U_s and U_p from experimental data**
- **On Hugoniot, the additional energy term should be zero**
- **Tantala experimental data by Vogler et. al. has initial densities at ~1.13, ~3.0, and ~7.4 g/cc**

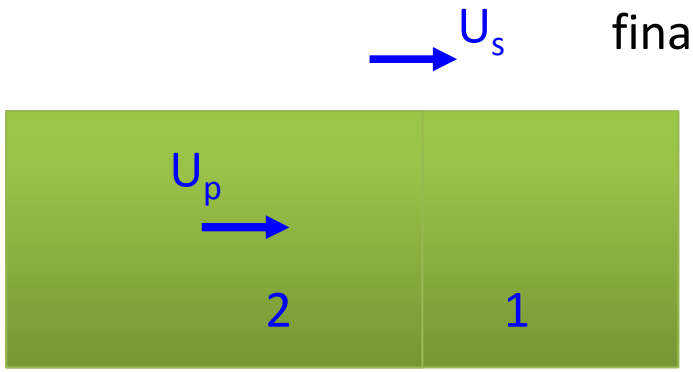




$P_{H,P}$ from Molodets, Combustion, Explosion, and Shockwaves, 42. 3. 2006

Shock compression is a way to investigate thermo-physical properties of matter at extreme pressures

- *Conservation of mass, energy, and momentum* lead to the **Rankine-Hugoniot condition** for the initial (1) and final state (2)



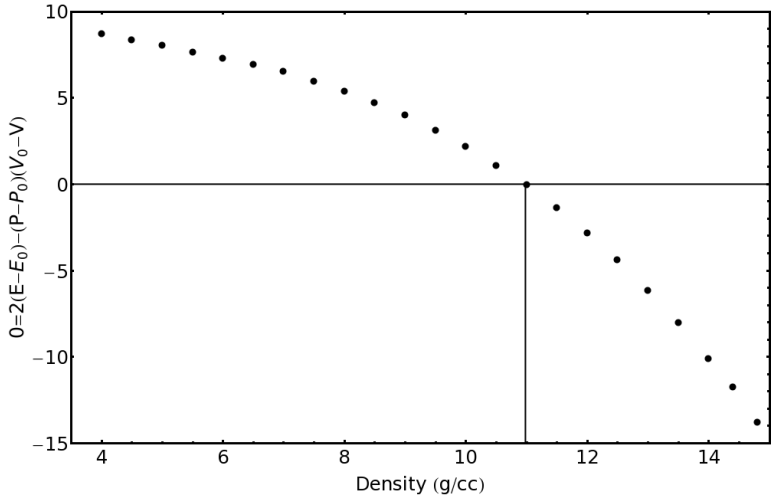
- E - internal energy
- P - pressure
- v – specific volume

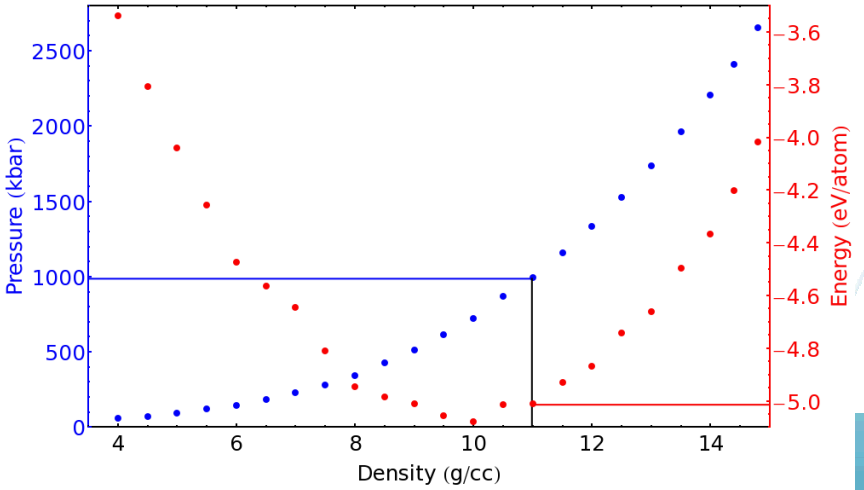
$$2\left(E_2 - E_1\right) = \left(P_2 + P_1\right)\left(\upsilon_1 - \upsilon_2\right)$$

- *Calculate the isentrope using the same jump relation but as a series of small shocks*

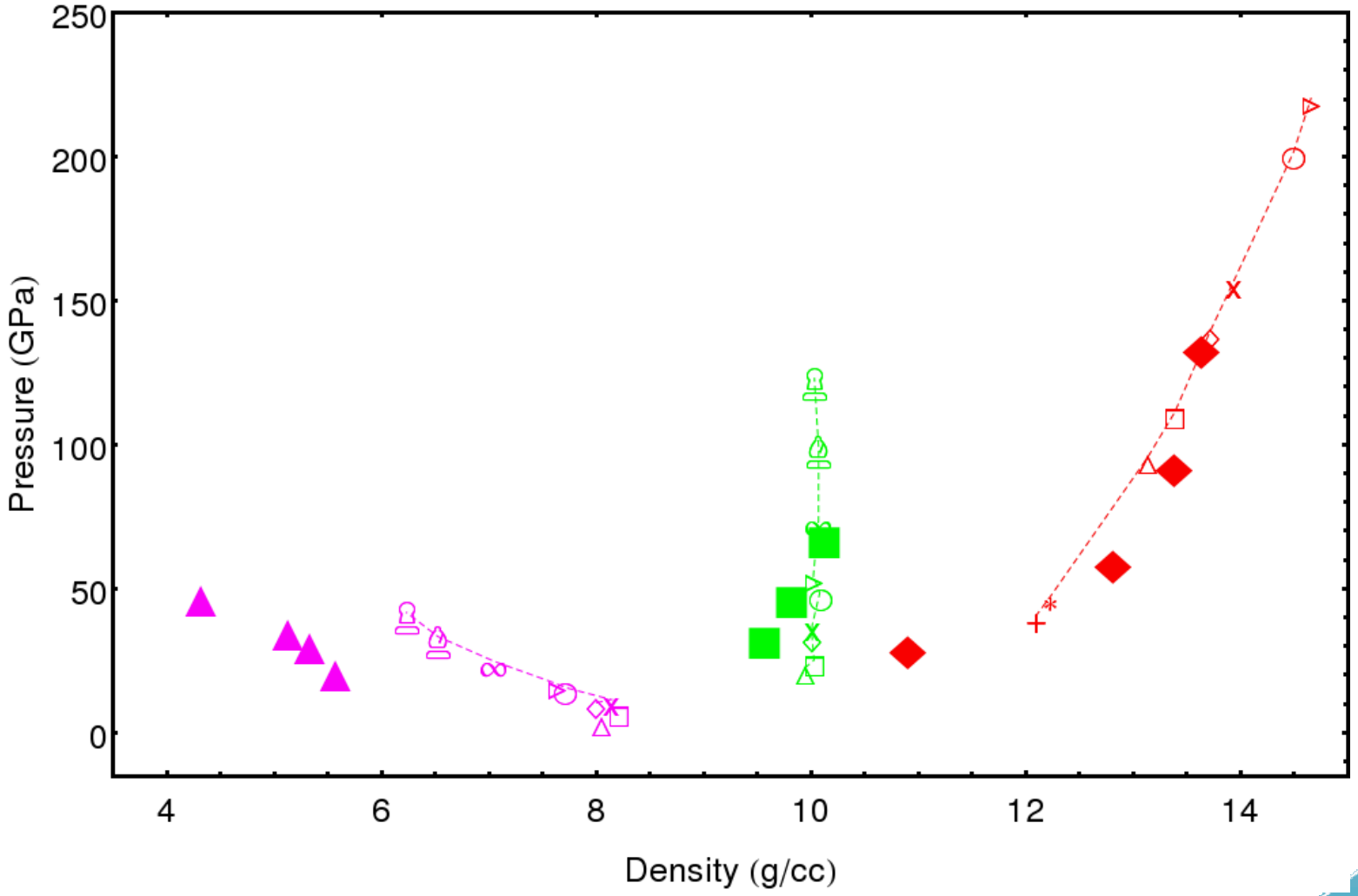
First-principles thermodynamics: use interpolation between points to obtain the Hugoniot

- **A series of equilibration simulations at constant temperature and different densities constitutes a DFT/QMD database**
- **For each temperature, we extract a pressure and energy profile**
- **The Rankine-Hugoniot relation is solved for each density/pressure/energy point**
  - **Interpolate in Rankine-Hugoniot space for density where relation is zero**
  - **Use this density to interpolate in pressure and energy space.**



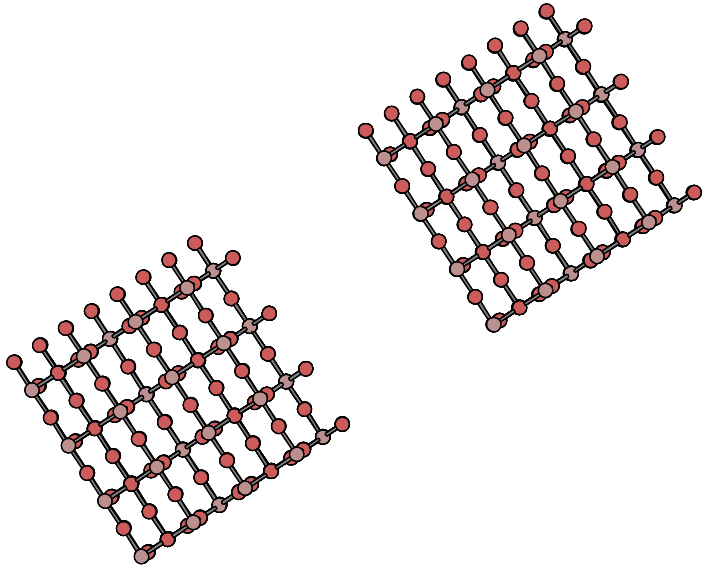


Comparison of VASP simulated and experimental Hugoniot points for tantala



The real systems are inhomogeneous: voids and flakes, it is necessary to take the surface energy into account.

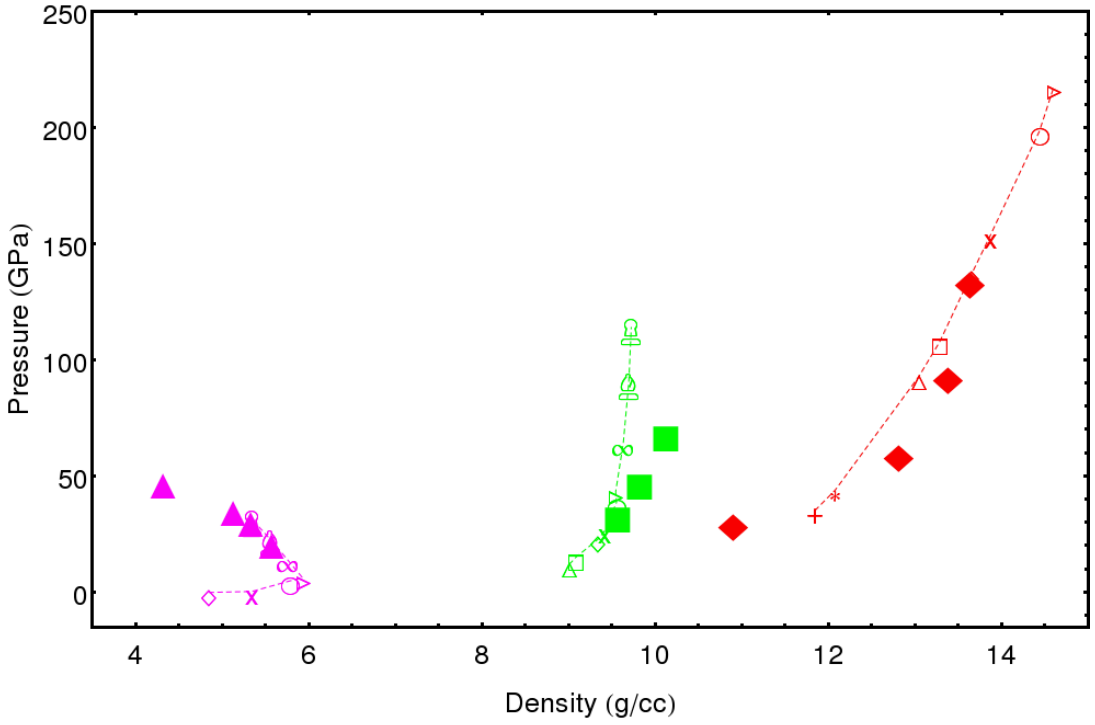
- **All of the initial densities start below solid (flakes and voids).**
- **Surfaces take more energy to maintain than internal bulk structure.**
- **Taking it into account in simulations**
  - **Create a basis cell that is large enough to contain internal and surface atoms but small enough to run**
  - **The energy reported is larger than the solid energy found in the earlier reference simulations**
  - **This difference is the "surface" energy.**
  - **Can also do single surface with multiple layers under with slightly different answers than a small block in void**



Simulation of a periodic system mimicking small flakes of tantala surrounded by void

Creating a Hugoniot plot based on many initial densities and including surface energy.

- The strategy for modeling shock compression of porous is accounting for a more complete energy balance of the Rankine-Hugoniot relation by including the surface energy
- δ=the difference between reference energy and surface energy
  - The same δ is used for all densities
- E_{solid} is the energy at the reference density (8.36 g/cc 300 Kelvin) from VASP
- P_{solid} is the pressure and usually NOT Zero (although experimentally it should be)
- V_0 is the starting experimental volume



$$0 = \left(E - E_0 - \delta\left[\frac{V_{00}}{V_0} - 1\right]\right) - \left(\frac{P + P_0}{2}\right)\left(V_{00} - V\right)$$

Analyzing the tantala experimental results in terms of an effective EOS - Mie Gruneisen Γ with surface energy corrections.

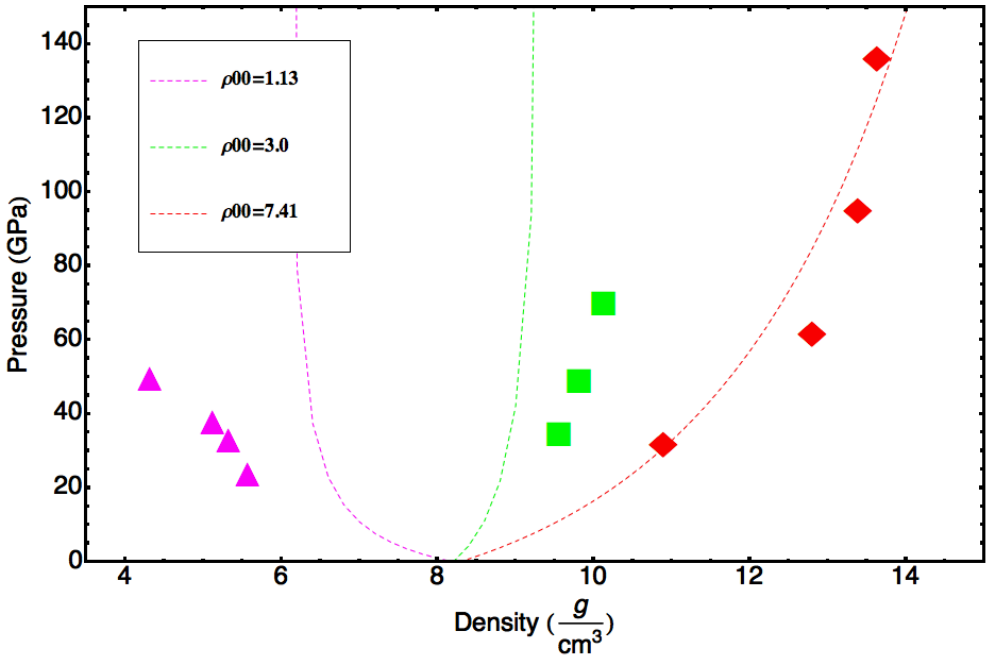
- **Added the surface energy term to the EOS corrects Mie-Gruneisen to more closely match experimental data**

$$P_H = \frac{\rho_o C_o^2 \chi \left[1 - \frac{\Gamma}{2}\chi\right]}{(1-\eta\chi)^2} + \Gamma\rho E$$

- **Where E is the off principal Hugoniot energy term and is of the form**

$$E = \delta\left[\frac{V_{00}}{V_0} - 1\right]$$

- **As can be seen, more work needs to be done on applying the surface energy for better EOS**



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Conclusions

- **We have developed an approach for simulating the Hugoniot for porous materials using first-principles methods (DFT/QMD)**

- **Accounting for the surface energy is required to get the porous Hugoniot correct**
  - **We formulated a ”Surface energy correction” inspired model to capture the effect**
  - **The method has inherent limitations, for example, polyethylene and TPX are materials with very small or no surface energy**

- **DFT/QMD was employed to calculate the Gruneisen Γ for tantala under extreme conditions**
  - **Γ is dependent on density and cannot be taken as constant**
  - **These calculations can be continued to obtain improved statistics**