High-Precision Measurements of the Equation of State of Polymers at 1 to 10 Mbar

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Precision equation-of-state (EOS) measurements are obtained on various polymers at 1 to 10 Mbar

- Precise knowledge of ablator EOS is required for ICF target designs
  - some NIF target designs use Ge-doped GDP

- Laser-driven shock waves produce EOS data using the impedance-matching (IM) method

- CH data allows for model discrimination, favoring SESAME 7592
  - mild softening is not accounted for between 2 to 4 Mbar
  - single- and double-shock results display similar behavior

- Stoichiometry effects between CH and CH\(_2\) are well-predicted by models

- EOS data for NIF ablator material was acquired
I. Motivation

II. Precision EOS measurements

III. Experiments

   A. Single shock, principal Hugoniot measurements
      i. Polystyrene (CH)
      ii. Polypropylene (CH$_2$)
      iii. GDP (CH$_{1.3}$O$_{0.023}$)
      iv. 0.6at% Ge-doped GDP

   B. Double shock, off Hugoniot measurements
      i. Polystyrene (CH)

C:H ratio

NIF ablators
Collaborators

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**Motivation**

High-pressure EOS data are required to understand high-energy-density (HED) physics.

- Reliable EOS data is important to dense plasma theory, where radiation hydrodynamic codes are used.
- Need material EOS over wide density and temperature ranges.
- Existing data covers a small fraction of these ranges.

EOS measurements above 1 Mbar are used to benchmark models.

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Some NIF ignition target designs use Ge-doped plastic ablators—high-pressure EOS measurements are needed.
Hydrocarbons are common ablator materials for ICF fuel pellets

- Ablator material properties are essential to the design and simulation of ICF targets
- By varying C to H ratio, the effect of stoichiometry on high-pressure behavior can be investigated

<table>
<thead>
<tr>
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<th>Formula</th>
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<tbody>
<tr>
<td>Polystyrene</td>
<td>CH</td>
</tr>
<tr>
<td>Polypropylene</td>
<td>CH$_2$</td>
</tr>
<tr>
<td>Glow discharge polymer (GDP)</td>
<td>CH$<em>{1.3}$O$</em>{0.023}$</td>
</tr>
<tr>
<td>Ge-doped GDP</td>
<td>CH$<em>{1.3}$O$</em>{0.023}$ + Ge$_{\text{at% 0.6}}$</td>
</tr>
</tbody>
</table>

EOS measurements on CH$_x$ will provide benchmark behavior on hydrocarbon polymers under extreme $P$, $\rho$ conditions.
Polymer EOS experiments were performed using laser-driven shock waves on OMEGA

- Experiments used laser energies between 200 to 1130 J delivered in a nominally 2-ns square pulse.
- Average laser irradiances on target were 0.3 to $1.1 \times 10^{14}$ W/cm$^2$

VISAR* has time resolution of <30 ps and shock-velocity precision of ~1%.
Impedance Matching $U_s = F(U_p)$

EOS data are obtained from the impedance-matching technique

Rankine–Hugoniot Equations

\[
\rho_0 U_s = \rho_1 (U_s - U_p)
\]

\[
P_1 - P_0 = \rho_0 U_s U_p
\]
Experimental errors must be minimized and systematic errors understood for precision EOS measurements.

- Measurement accuracy depends on knowledge of standard.
- Most impedance-matching (IM) studies quote only random errors.
- Cannot propagate systematic errors using theoretical EOS.

- Random errors

\[
\frac{\delta \rho}{\rho} \propto (\eta - 1) \times \frac{\delta U_s}{U_s}, \quad \text{where} \quad \eta = \frac{\rho}{\rho_0};
\]

\[
\eta \approx 4 - 6 \rightarrow \frac{\delta \rho}{\rho} \propto (3 - 5) \times \frac{\delta U_s}{U_s}
\]
Random Errors

Higher precision is obtained with a transparent standard compared to an opaque standard.

**Random Errors**

- Only information is transit time
- Can use only integrated shock
- No knowledge of shock stability

**EOS observables** are obtained at the contact interface.

**Sample**

VISAR-1 shot 29425

- Contact interface
- $U_s$ is inferred from transit times
- Integrated velocity

VISAR-1 shot 52118

- $\alpha$-quartz pusher
- Instantaneous velocities
- Contact interface

$U_s$ is inferred from transit times.
Higher precision is obtained with a transparent standard compared to an opaque standard.

Random Errors

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- No knowledge of shock stability

**Sample**

VISAR-1 shot 29425

Integrated velocity

VISAR-1 shot 52118

Contact interface

\( U_s \) is inferred from transit times

\( \Delta x \)
α-quartz has been validated as an EOS standard

Systematic Errors

α-quartz EOS (Al as reference)

Mbar 1.4 4.5 9.45 15.94

Shock speed (μm/ns)

35 30 25 20 15 10 5

Particle speed (μm/ns)

Laser¹ ~0.3 ns
Nuclear ~10 μs
Gas gun explosive ~100s of ns

SiO₂ Aerogel

Release isentrope (±)
Direct impact

Pressure (Mbar)

Direct measurement²
IM with Al standard²
IM with α-quartz standard³

\( \alpha \)-quartz’s release isentrope is approximated using the Mie-Grüneisen EOS

- \( \Gamma \) describes pressure differences between equal volume states on the Principal Hugoniot

\[
\Gamma = V \left( \frac{dP}{dE} \right)_v
\]

- Combining the above with the first law of thermodynamics,

\[
dE = T dS - P dV
\]

with \( dS = 0 \), leads to a recursion relation describing a loci of isentropes in the \( P-V \) plane

- Based on models, \( \Gamma \) is assumed to be constant in the high-pressure fluid regime, with value \( \Gamma = 0.64 \pm 0.11 \)
Precision EOS data tightly constrain polystyrene (CH) EOS models

- SESAME 7592 provides the best fit to the measurements
- CH displays softer behavior between 2 to 4 Mbar than SESAME 7592

Gas gun data

Total error

Random error

Softening?
The dependence on the C:H ratio is well-predicted by models
The polystyrene results have higher precision than previous studies.

Shocked CH and CH$_2$ become reflective at 1 to 2 Mbar

• Reflectivity measurements are needed for temperature calculations

Expected behavior of dielectrics undergoing insulator-conductor transition.
The measured brightness temperatures are consistent with models; but differences among models are too small to be discerned.

This provides a complete EOS of CH and CH₂.
Preliminary data on Ge-doped GDP displays softer behavior than most models.
Double Shock

Shock transit into a higher impedance material results in a reflected shock.

**Graphical Representation**

- **Sample** and **Standard** materials are depicted with different pressures and particle velocities.
- **Intersection** yields the pressure and particle velocity in the sample.
- **Possible shock states** in the sample are indicated.
- **Reshock** and **Initial state** are marked.

**Equation**

- \( P = \rho_0 U_s U_p \)
- \( P = \rho_0 U_s' U_p \)

**Legend**

- **Sample principal hugoniot**
- **Pressure** (Mb)
- **Particle velocity, \( u_p \) (\( \mu m/\text{ns} \))
Reflected shocks are used to create double shock states in CH

Small differences in models are amplified using reshock to move off the Hugoniot.
Polystyrene (CH) double-shock data are in agreement with single-shock results
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Inclusion of a softer $\alpha$-quartz EOS produces ~0.2% to 6.0% difference in polystyrene density values.