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Precision Equation-of- State Measurements on NIF ablator materials from 1 to 12 Mbar Using Laser-Driven Shock Waves

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Abstract

A large uncertainty in the design of ignition capsules for use in the National Ignition Campaign (NIC) is the ablator equation of state (EOS). In this article we report equation-of-state measurements for two candidate NIC ablator materials, glow-discharge polymer (GDP), and germanium-doped GDP (Ge-GDP). These materials were driven to pressures of 1 to 12 Mbar using laser-driven shock waves. Hugoniot measurements were obtained using the impedance matching technique with an α-quartz standard. This article presents the first kinematic measurements in the high-pressure fluid regime for these

materials, which show to be in close agreement with Livermore Equation-of-State (LEOS) model predictions.

I. INTRODUCTION

Hydrocarbons are often used as ablator materials for inertial confinement fusion (ICF) studies, satisfying low-surface-roughness constraints and enabling one to introduce dopant atoms that are chemically bonded to the hydrocarbon, preventing its migration through the capsule shell.[1] One of three indirect-drive capsule ablator designs for the National Ignition Facility (NIF)[2] calls for glow-discharge polymer ($CH_{1.3}O_{0.02}$) (GDP) ablators with various levels of germanium doping (Ge-GDP).[1,3–5] Introducing a mid-*Z* dopant reduces preheating of the fuel by increasing the opacity to hard x rays (M bands) generated from the hohlraum wall; it also lowers the Atwood number.[1,5] This helps to optimize target performance and relaxes manufacturing constraints.

Ultimately, each proposed NIF target design has advantages and weaknesses. The choice of ablator relies on detailed hydrodynamic simulations to specify the shell dimensions, dopant levels, and laser pulse. The aim is to produce a sufficiently robust design such that ignition will be achieved despite experimental uncertainties and hydrodynamic instabilities. These simulations require knowledge of the ablator equation of state (EOS), setting constraints on tolerable capsule surface roughness, ablator thickness, and driver energy. Instability growth rates have been shown to strongly depend on the ablator compressibility and first-shock strength as defined by its EOS.[1,6]

The results presented here form part of a larger study that aims to better understand the behavior of hydrocarbon ablators and characterize the high-pressure

behavior of germanium-doped GDP for NIF ignition targets. Initially there was concern that variation in material properties among GDP and Ge-GDP batches could compromise the results. With this in mind, the EOS of CH (polystyrene) and CH_2 (polypropylene), two materials whose fabrication process is well known and reproducible, were measured to high precision.[7] This resolved uncertainties in the high-pressure (≥1-Mbar) behavior of CH and provided high-pressure measurements of the effect of changes in stoichiometry by varying the H-to-C ratio through EOS measurements of CH_2. These results provided the basis for benchmarking interim tabular models developed for GDP and Ge-GDP used for ignition target designs.

Up to now, the EOS of CH was used as a surrogate EOS to model GDP ablators. In comparison to CH, however, GDP has a higher H-to-C ratio and trace amounts of oxygen, absorbed by the films during the fabrication process. Material properties such as initial density and index of refraction differ between CH and GDP/Ge-GDP films. Although the EOS of CH is known to high precision,[7] it was nevertheless important to study the behavior of GDP and Ge-GDP with equal precision. This is to verify how differences in stoichiometry and material properties influence the EOS of such materials and compare with model predictions.

This study presents the first EOS measurements of GDP and Ge-GDP up to pressures of ~10 Mbar, driving these materials into the high-pressure fluid regime. These experiments used laser-driven shock waves to provide impedance-matching (IM) conditions with α-quartz used as the standard material.[8,9] Previous studies used quartz as a standard with successful results, demonstrating a significant reduction in measurement uncertainties.[7,9,10] By using velocity interferometry and transparent materials, ~1%

precision was obtained in shock-velocity measurements.[11] GDP and Ge-GDP data are compared with available LEOS (Livermore equation of state) models, showing close agreement between measurements and model predictions. The IM technique, its associated error analysis, and optimization-enabling precision measurements are discussed in Sec. II. The experimental configuration, targets, and diagnostics are described in Sec. III, followed by results in Sec. IV, and concluding remarks in Sec. V.

II. IMPEDANCE-MATCHING TECHNIQUE

The impedance-matching technique allows one to determine a sample's particle velocity (U_p), pressure (P), and density (ρ) through shock-velocity measurements in a reference material, here z-cut α-quartz ($U_{s,Q}$), and the sample being tested, i.e., GDP ($U_{s,GDP}$) or Ge-GDP($U_{s,Ge\text{-}GDP}$).[12,13] Measurement of the shock velocities in the quartz and the sample at the contact interface yields the initial shock state from which the quartz releases and the shocked state in the sample. The Rankine–Hugoniot relations are met at the shock front at all times and allow determination of the stress and density of the sample.

In this study, the IM analysis was performed using quartz's experimentally determined Hugoniot[8,14] and a constant Grüneisen parameter (Γ) to calculate off-Hugoniot release states.[7,14,15] Using quartz (ρ_0 = 2.65 g/cm^3, n = 1.547) as an IM standard provides higher precision than opaque standards by reducing errors associated with the standard's initial state, from which off-Hugoniot curves are launched. It relaxes the shock-stability constraints since changes in shock velocity are directly measured and

can be accounted for in the analysis. Measuring *in-situ* shock velocities permits one to measure observables ($U_{s,Q}$, $U_{s,GDP}$, and $U_{s,Ge\text{-}GDP}$) close to the contact interface, where the IM conditions are valid.

The data precision obtained through the IM technique is strictly correlated to the accuracy with which the states in the standard are known. Systematic uncertainties enter the IM analysis through calculation of the standard's Hugoniot and off-Hugoniot states. Quartz's shock EOS was previously measured in the pressure range ~2 to 15 Mbar through impedance matching with an aluminum (Al) standard, using laser-driven shock waves.[8] These experiments were in agreement with previous gas-gun and nuclear and chemical explosive studies, displaying linear behavior in the U_s–U_p plane, given by a piecewise linear function of general form $U_s = a_0 + a_1(U_p - \beta)$,

$$U_s = (6.91 \pm 40.0) + (1.66 \pm 70.0)(U_p - 1.865), \text{ for } U_p < 6.358\ \mu\text{m/ns}, \quad (1)$$

$$U_s = (19.501 \pm 0.068) + (1.276 \pm 0.022)(U_p - 11.865), \text{ for } U_p \geq 6.358\ \mu\text{m/ns}, \quad (2)$$

where an orthonormal basis is used such that resulting errors are uncorrelated.[14] This fit was used in this study's analysis.

Quartz's release states are approximated by a Mie–Grüneisen EOS as described in McQueen *et al.*[15] Based on solid and porous silica Hugoniot measurements,[8,14,16,17] quartz's Grüneisen parameter was shown to be nearly constant with $\Gamma = 0.66 \pm 0.1$. Model predictions in a similar pressure range estimate the value to be $\Gamma = 0.64 \pm 0.11$ (Ref. 9), in

agreement with experimental results. This analysis uses the model-based value for Γ, the only model-based parameter entering this IM analysis. Using the quartz experimental principal Hugoniot and a Mie-Grüneisen formalism to approximate release states enables one to propagate systematic uncertainties inherent in the IM technique.

Random uncertainties enter the IM analysis through calculation of the quartz and sample Rayleigh lines, $P = \rho_0 U_s U_p$, describing the thermodynamic path taken during compression in the P–U_p plane. These errors are often only due to shock-velocity measurement uncertainties; in the case of GDP and Ge-GDP, uncertainties in their initial density were also accounted for, translating to an increase in random errors compared to previous precision EOS measurements.[7] The total error associated with the measured U_p, P, and ρ corresponds to the quadrature sum of random and systematic uncertainties.

IM calculations are often performed in the P–U_p plane since both of these quantities must be continuous across the contact interface between the standard and sample. In the previously described framework, the final (P, U_p) is a function of nine parameters: a_{0L}, a_{0H}, a_{1L}, a_{1H}, Γ, ρ_{0Q}, ρ_{0CHx}, $U_{s,Q}$, and $U_{s,CHx}$, and their corresponding uncertainties.[7] The first four variables are fitting parameters for the quartz principal Hugoniot, where subscripts L and H refer to the low ($U_p < 6.358$ μm/ns) and high ($U_p \geq 6.358$ μm/ns) fits. The fifth parameter corresponds to the model-dependent Grüneisen parameter, and the last four correspond to the quartz and sample initial density and measured shock velocity. Because quartz's experimentally derived principal Hugoniot and Grüneisen-based release can be considered independent, error contributions from each parameter are found by calculating deviations from the nominal U_p, P, and ρ results.

Note that this amounts to eight error contributions since no error is assumed for quartz's initial density.

Subsequent work at the Sandia Z Machine on the EOS of quartz in the 1- to 16-Mbar pressure regime[18] observed curvature in the U_s–U_p plane not present in the EOS used herein. If real, this difference in EOS would cause systematic shifts in the derived sample EOS. Although these discrepancies in the principal Hugoniot of quartz are yet to be resolved, it is important they are acknowledged as they apply to the IM analysis. This subject is discussed in Appendix A.

III. EXPERIMENTAL METHOD

Experiments were performed on the University of Rochester's OMEGA laser,[19] a frequency-tripled Nd:glass laser that produces 351-nm light. Shock pressures were generated using ~250 to 2100 J delivered in a nominally 2-ns square pulse. Focal-spot profiles were smoothed using distributed phase plates,[20] leading to a uniform irradiation area with a 600- or 800-μm diameter. This resulted in average irradiances on target of 0.24 to 2.5 $\times 10^{14}$ W/cm^2.

Experiments were conducted using multilayered 3-mm $\times$ 3-mm planar targets, which had a 20-μm CH ablator to prevent preheating of the target by minimizing x rays generated in the coronal plasma and a 90-μm-thick pusher made of *z*-cut α-quartz, used as the IM standard. On the rear side of the pusher were two samples: ~30 μm of Ge-GDP on the top half of the target and ~30 μm of GDP on the bottom half of the target. This allowed simultaneous EOS measurements of both materials.

Ideally the probe beam used to diagnose shock velocities only reflects off the moving shock front; in practice, the system can register back-reflections and internal reflections from the target layers, referred to as "ghost fringes." To mitigate these effects, the free surface of all targets, those opposite the drive beams, had an antireflection coating to minimize ghost reflections. Because IM measurements are performed at the contact interface between the standard and sample, glue layers between these materials were kept at a minimum, estimated to be no more than 1 to 2 μm thick. For some shots the GDP and Ge-GDP films were directly deposited onto the α-quartz, requiring no glue layer.

GDP and Ge-GDP films were made and characterized by General Atomics (GA)[21] in small batches. The material properties of these films, such as stoichiometry, initial density, and index of refraction, varied slightly among batches. These differences were taken into account in the data analysis and are summarized in Table I. Elemental compositional stoichiometry was based on x-ray fluorescence (XRF) chemical analysis. Average initial densities were obtained by measuring the volume and weight of each film batch. The film length and width were measured within 0.1 mm, and thicknesses were measured by interferometry techniques to within 0.5 μm, resulting in initial density uncertainties between 1% and 2%. For each batch, a separate sample fabricated from the same batch material was used to measure index of refraction. This sample was submerged in various Cargille Laboratories[22] series-A index-matching fluids. White-light interferometry with a 532-nm filter was used to determine the best match among the various index-matching fluids. The index of refraction for the matching fluid was verified using an Abbe refractometer, finding agreement to ±0.0005. Both the Abbe refractometer

and the index-matching fluids are referenced to the sodium D line at λ = 589.3 nm (at 25°C). The Cauchy equation, with coefficients provided by the vendor, were used to find the index of refraction at λ = 532 nm. Uncertainties associated with index-of-refraction measurements arise from the estimated fringe offsets and wavelength correction with the exception of one batch the index of refraction for which GDP was measured to be 1.571±0.005 and Ge-GDP was measured to be 1.572±0.005 (see Table I).

Shock velocities were measured using a line-imaging velocity interferometer system for any reflector (VISAR),[11] with an 800-μm field of view. Two interferometers with different sensitivities were used to discern the 2π phase-shift ambiguity occurring at fringe jumps and breakout times. Uncorrelated velocity sensitivities of 2.732 and 6.906 μm/ns/fringe were produced by using etalons with 18- and 7-mm thickness, where the velocity sensitivity in each material varied based on its index of refraction. The dielectric materials used were subject to drive pressures in the Mbar range, generating reflective shock fronts. The VISAR probe laser—a *Q*-switched, injection-seeded Nd:YAG laser operating at 532 nm with an ~50-ns full width at half maximum (FWHM), reflected off the shock front. This signal was relayed to the set of interferometers and recorded on streak cameras with a 15- or 9-ns temporal window (one for each VISAR), providing streak images with temporal and 1-D spatial resolution. Since the shock fronts were highly reflective, the measured Doppler-shifted signal detected by VISAR is directly correlated to the velocity of the shock wave. Streak images were post-processed using a fast Fourier transform (FFT) method,[11,23] resolving fringe jumps to ~5% of a fringe. This translates to a ~1% precision in shock-velocity measurements, with typical measurements resulting in five fringe shifts. The streak cameras had temporal resolutions

close to 10 ps and the etalon delay times used in the interferometers were either 40 or 90 ps.

The experimental configuration is shown in Fig. 1(a); the OMEGA[19] beams irradiate the front of the target, with VISAR aligned opposite the drive beams perpendicular to the rear target surface. Because targets were composed only of transparent materials, VISAR provided a continuous shock-velocity history as the shock wave transited each target layer. Figure 1(b) shows a section of the VISAR streak image, corresponding to the bottom half of the target (GDP). Here the quartz was driven to a shock pressure of 8.6 Mbar, corresponding to 4.9 Mbar in GDP and 5.1 Mbar in Ge-GDP. In Fig. 1(b) the laser drive starts at $t = 0$ and the shock enters the quartz at ~0.6 ns. By ~2 ns, the shock in the quartz equilibrates with the pressure caused by material ablation, considerably stabilizing its velocity. At ~4.5 ns the shock reaches the GDP sample and reaches the sample's rear surface at ~6 ns. It is clear from Fig. 1(b) that the quartz:GDP contact interface has finite temporal width (at 4.5 ns) due to the glue layer and VISAR temporal resolution of 40 or 90 ps. (The same is true for the quartz:Ge-GDP interface, not shown.) Consequently, shock velocities are not measured at the "true" contact interface between reference and sample, but at an earlier and later time, respectively. This is accounted for by linearly fitting the velocity profile over ~300 ps and extrapolating to the time where the true contact interface would be, here defined to be the equidistant (temporal) location between the quartz and sample boundary, shown as t' in Fig. 1(c). This methodology accounts for any deceleration observed in the measurements, such that the IM conditions are still matched despite shock-wave decay. Figure 1(c) shows the resulting shock-velocity history for quartz and GDP extracted from the streak image data

shown in Fig. 1(b) after applying the FFT to the image and matching velocity solutions from both interferometers.

IV. RESULTS AND DISCUSSION

A. Glow-discharge polymer

Single-shock principal Hugoniot measurements for GDP were obtained from ~1 to 12 Mbar. The experimental observables and resulting kinematic properties for GDP are listed in Table II. The results display a centered linear U_s–U_p relation, as shown in Fig. 2, given by U_s = (24.56±0.07) + (1.29±0.01) (U_p–16.76). The average particle velocity $\overline{U_p} = 16.786$ such that the fitting coefficients are independent and the uncertainties are uncorrelated.[24] Because of this independence, the errors associated with such fitting parameters are independent. The initial density, compositional stoichiometry, and index of refraction of the samples varied between fabrication batches (Table I); these variations were included in the analysis. Initial material densities are distinguished in Fig. 2 by color shades as ρ_0 = 1.06 g/cm^3 (dark green diamonds), 1.05 g/cm^3 (medium green diamonds), and 1.044g/cm^3 (light green diamonds). Measurement uncertainties in the U_s–U_p plane are within ~1%. Alterations of the high-pressure behavior caused by variations in initial properties were not evident in the U_s–U_p plane; therefore, no distinction was made among the batches for the linear fit in this plane. The data is shown to be in agreement with GDP LEOS model 5310.

Current LEOS models are developed based on the quotidian equation of state (QEOS)[25]–type construct, where an additive approach is taken to describe the Helmholtz

free energy accounting for ion/nuclear and electron contributions with semi-empirical bonding corrections and/or quantum/exchange corrections if needed. Typically, the electron contribution is calculated via a Thomas–Fermi model and the ion contribution via the Cowan EOS,[25,26,27] where equations are parameterized such that limiting values (at solid density, as ρ approaches ∞, etc.) yield reasonable results and experimental data is recovered.[25] Because experimental EOS measurements for GDP were unavailable at the time LEOS 5310 was constructed, this model was benchmarked[1] using precision EOS measurements for CH[7] in the high-pressure regime and previous CH gas-gun data at lower pressures,[28] where density scaling based on the equilibrium densities of CH and GDP was used where applicable.[27] The modeled GDP behavior was further constrained by generating data from CHEETAH, a thermochemical model.[27] LEOS 5310 does not include dissociation. The assumed initial density and stoichiometry for LEOS 5310 are listed in Table III.

The H-to-C ratio and initial density of LEOS 5310 were slightly different than the GDP samples used in these experiments; differences varied by ~1%, on average, for both parameters. Oxygen levels measured in the GDP films were quite small (O-to-C ratio of 0.01 and 0.02) and had no detectable effect on the material's high-pressure behavior since batches with different oxygen levels compared well in both the U_s–U_p and P–ρ planes (see Figs. 2 and 3). The oxygen doping in LEOS 5310 model is comparable to that found in the films.

Figure 3 shows GDP results in the P–ρ plane, with data colors as previously described [ρ_0 = 1.06 g/cm^3 (dark green diamonds), 1.05 g/cm^3 (medium green

diamonds), and 1.044g/cm^3 (light green diamonds)]. Here the LEOS 5310 model was evaluated with initial density ρ_0 = 1.05 g/cm^3, the average initial density between the GDP batches. *SESAME* 7592 (Ref. 29), the CH *SESAME* model that best fit the precision polystyrene data, is shown as a reference. The data are in close agreement with LEOS 5310 model predictions and are only slightly softer than *SESAME* 7592, showing that GDP behaves quite similarly to CH in the high-pressure fluid regime. In this plane it is also evident that data precision is degraded in comparison to previous precision EOS studies, yielding 3.4% to 5.6% precision in density. This is due to the inclusion of initial density uncertainties, which accounted for 5.2% to 44% of the total density error.

B. Germanium-doped GDP

Germanium-doped GDP (Ge-GDP) was studied in the ~1- to 12-Mbar pressure range. Table IV presents shock-velocity measurements and kinematic properties obtained through the IM technique using quartz standard. With the exception of one batch, the initial density and index of refraction of the Ge-GDP films were consistent (Table I), although levels of Ge doping varied from 0.5% to 0.7%. The equation of state of Ge-GDP in the U_s–U_p plane (shown in Fig. 4) displays a centered linear relation given by U_s = (23.39±0.07) + (1.31±0.01) (U_p–15.92). Again, the errors are independent. In this figure Ge-GDP data are compared to available models for Ge-doped GDP, LEOS 5312, and LEOS 5315 with assumed 0.2% and 0.5% Ge doping. These models display a similar behavior in this plane and are in agreement with measured data. LEOS 5312 and 5315 models for Ge-GDP were developed in a similar fashion as previously described for LEOS 5310. Because CHEETAH was not equipped to model Ge at the time, Si was used

instead. LEOS 5315 and 5312 were developed independently; therefore the fit to the CHEETAH model is different for these models (LEOS 5310 and 5315 share the same fit). This is evident in the P–ρ plane (Fig. 5), where the models cross at ~2 Mbar. Like LEOS 5310, LEOS 5312 and 5315 do not include a dissociation model.[27]

Development of LEOS models for GDP and Ge-GDP is an ongoing effort. Much of the difficulty in developing models that accurately describe material behavior over a wide range of states is a lack of experimentally measured material properties, which need to be inferred through codes such as CHEETAH or approximated using measured quantities of similar materials. Future LEOS table series, sharing the same fit to CHEETAH results, will include a dissociation model, have more stringent constrains to release states, and include new material-response measurements as they become available, including this work. Material properties assumed by LEOS 5312 and 5315, such as initial density and compositional stoichiometry, are listed in Table III.

The total uncertainty in density was between 3.5% and 5.4%. Isolating random errors due to Ge-GDP film initial density uncertainty ($\delta\rho_{0\mathrm{GeGDP}}$) and comparing to the total density error, representing the quadrature sum of random and systematic uncertainties, it was noted that $\delta\rho_{0GeGDP}$ contributed anywhere between 7.2% to 40% of the total density uncertainty (varying as a function of pressure). Figure 5 shows Ge-GDP EOS results in the P–ρ plane. Here samples with initial density of 1.13 g/cm^3 are shown as purple circles (Ge at 0.7%), diamonds (Ge at 0.7%), and squares (Ge at 0.5%), while data taken on samples with initial density of 1.10 g/cm^3 are shown as dark purple squares (Ge at 0.5%). The random errors are shown as the colored error bars, which included shock velocity (in Ge-GDP and quartz) and initial density uncertainties. Total errors are

shown as gray error bars. Data are compared with LEOS 5312 and LEOS 5315, both evaluated at an initial density of 1.13 g/cm^3. LEOS 5310, evaluated at ρ_0 = 1.05 g/cm^3, and *SESAME* 7592 are plotted on this figure as a reference. On average, the percent difference between the model and sample H-to-C ratio was ~3.5%, although it varied up to 5.6%, and oxygen levels assumed in the models were smaller than those measured. Despite these differences, Fig. 5 shows that the data follow similar trends and agree with model predictions to within experimental errors. Between 2- and 4-Mbar, a slight softening of the data compared to model predictions is observed. Above these pressures, the Ge-GDP data begin to stiffen (becoming less compressible) approaching closer agreement with theoretical models. Although differences among these models are amplified in the P–ρ plane, the data presented here are not able to discriminate among models due to the random uncertainties.

Plotting the Ge-GDP results in the pressure–compression (P–ρ/ρ_0) plane (see Fig. 6) shows a smooth trend in close agreement with LEOS models for all pressures, i.e., no stiffening as observed in the P–ρ plane. The apparent stiffness at higher pressures observed in the P–ρ plane results from initial density variations. Ge-GDP was observed to undergo compression between 2.5× and 3.4×, comparable to those reached by GDP, as shown in Fig. 7. Even with the fairly high precision of these measurements, errors in the data (including the initial density uncertainty) make it difficult to favor any model behavior in this plane, even at the highest pressures, where models diverge most.

V. CONCLUSIONS

The high-pressure behavior of GDP and Ge-GDP was measured at shock pressures of ~1 to 12 Mbar. The use of quartz as the IM standard along with a time-resolved VISAR diagnostic facilitated measuring shock velocities to ~1% precision in transparent materials. This reduced the uncertainties in the initial shock state of quartz, minimizing measurement errors in the GDP and Ge-GDP EOS results. Using quartz's experimentally derived principal Hugoniot[8] and a Mie–Grüneisen EOS, systematic uncertainties inherent in the IM technique were accounted for in the analysis. Density uncertainties of 3.4% to 5.6% (GDP) and 3.5% to 5.4% (Ge-GDP) incorporate initial density variations in the samples. The initial density error contribution varies as a function of pressure and estimated error value, contributing up to 40% of the total error in the GDP and Ge-GDP density measurements.

The GDP results are in close agreement with the LEOS 5310 model in both the U_s–U_p and P–ρ planes, despite differences in stoichiometry of ~1%. Likewise, the data indicate that GDP behaves similarly to CH when shocked to greater than ~1 Mbar, displaying only a slight softness at higher pressures in comparison to this material. The effect of germanium doping on GDP samples is predicted reasonably well by available LEOS models, which are in agreement with measured Ge-GDP results, despite differences in assumed material stoichiometry. The compressibility of Ge-GDP on the Hugoniot is well predicted by both LEOS 5312 and LEOS 5315, although the data cannot discriminate between the two models.

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APPENDIX A: INFLUENCE OF QUARTZ AS A STANDARD

Impedance-match results are relative measurements since the sample's properties are referenced to the material response of the standard. Recent experiments performed on the Sandia Z Machine by Knudson *et al.*[18] observed noticeable curvature in its $U_s(U_p)$ relation, previously observed to be linear in laser experiments.[8] As a result of this discrepancy there is an uncertainty in the measured behavior of GDP and Ge-GDP, as well as all other materials whose properties were measured through IM with a quartz standard. We present here an assessment of the effects of using a different quartz EOS, were the Knudson result to be accurate.

The EOS of quartz enters the IM analysis through the principal Hugoniot and the off-Hugoniot (release) states. Because of the nature of the IM technique, differences in the standard's EOS will propagate through and get amplified through the analysis. The analysis presented in this study approximates release states using a Mie–Grüneisen EOS (this assumes Γ is independent of density such that Γ/V is constant) with $\Gamma = 0.64$. It is yet to be determined if the observed quartz behavior by Knudson *et al.*[18] would still be

consistent with such Grüneisen parameter value. It is therefore important to assess the degree of discrepancy among IM results that would arise as a consequence of applying a different standard EOS.

To compare both fits on an equal basis, the reflected-Hugoniot approximation was used to arrive at IM results, using measured quantities from CH, CH_2, GDP, and Ge-GDP experiments. Although the reflected Hugoniot is not a highly accurate approximation for these pressures and material properties, the interest here is in the *differences* among states as a result of applying a given quartz EOS. The percent differences in the CH, CH_2, GDP, and Ge-GDP density and pressure are shown in Fig. 8. Here a positive percent difference indicates the quartz fit obtained through laser data leads to larger pressure or density values, in comparison to the Z-machine quartz fit. The dashed blue and solid pink curves represent the percent differences in density and pressure between the two quartz fits, peaking at ~4% and 6% in pressure and density. The orange, light blue, green, and purple circles and squares represent the percent differences in density and pressure for CH, CH_2, GDP, and Ge-GDP, respectively. This analysis incorporates deviations in quartz's initial shock state and calculated release curves between laser and Z-machine quartz fits. Because these materials have similar impedances, the percent differences follow similar trends as a function of pressure (shock velocity), peaking at comparable quartz shock velocities. For these materials the maximum deviations in pressure and density were found to be ~4.4% and 8.8% at quartz shock velocities of 18 to 19 μm/ns, corresponding to hydrocarbon pressures of ~3.5 Mbar. At hydrocarbon pressures below (above) ~2 Mbar (~8 Mbar) percent differences in density are within measurement error.

Systematic uncertainties between the two quartz fits can be compared through inferred single-shock states from double-shock measurements. This inversion method was previously described by Hicks *et al.*[30] and was applied to double-shock CH measurements, yielding inferred single-shock states in agreement with single-shock measurements.[7] This method uses a model-averaged effective gamma to calculate off-Hugoniot states (in this case the CH re-shock), using the quartz fit only to establish the double-shock particle velocity and pressure. The percent differences resulting from different quartz models are shown in Fig. 8 as the red circles (density) and squares (pressure). This method yields percent differences in pressure and density peaking at 3.2% and 6.3%. Inferred states using laser and Z-machine quartz EOS agree within measurement uncertainties below ~2.7 Mbar and above ~7.1 Mbar. This corresponds to a larger range of agreement than that predicted by the reflected Hugoniot analysis. Here quartz's influence enters only through the double-shocked pressure and particle velocity in CH.

Understanding the behavior of Γ under the Z-machine quartz EOS is equally important to the IM analysis. Having a different constant value or one that is non-constant and dependent on density will influence the calculated release states in quartz and result in different IM states. This issue has yet to be resolved.

The use of quartz as a standard in the IM technique has greatly improved the precision of EOS measurements, demonstrating the ability to provide EOS data that is able to more tightly constrain the high-pressure behavior of materials.[7,9] A new equation of state of quartz could generate slightly different final EOS results but would not compromise the precision of these data.

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FIGURE CAPTIONS

FIG. 1. (a) (Color online) Schematic of two material targets used in the experiments. Here the OMEGA beams irradiate the target at 23° and 48° to target normal, with VISAR aligned and focused to the rear surface of the target. Multilayer planar targets were composed of a CH ablator, quartz standard, and two different samples (GDP and Ge-GDP) to be tested. (b) VISAR streak image corresponding to the bottom half of the target, containing data for GDP, shows continuous tracking of the shock front within the standard and material being tested. (c) Final shock-velocity measurements in quartz and GDP after data extraction and analysis. For the IM analysis, shock velocities are evaluated at t'.

FIG. 2. (Color online) Principal Hugoniot measurements and models for GDP in U_s–U_p plane. Data were obtained through the IM construct with quartz reference on GDP films with initial density ρ_0 = 1.06 (dark green diamonds), 1.05 (medium green diamonds) and 1.044 (light green diamonds) g/cm^3.

FIG. 3. (Color online) Principal Hugoniot data and models for GDP in P–ρ plane, with GDP film's initial densities as previously described. Data are compared with CH *SESAME* 7592 (Ref. 30) and LEOS 5310 model, evaluated at initial density ρ_0 = 1.05 g/cm^3. Random uncertainties are shown as green (smaller) error bars and total uncertainties (quadrature of random and systematic uncertainties) as the gray (larger) error bars.

FIG. 4. (Color online) Principal Hugoniot measurements and models for Ge-GDP in U_s–U_p plane. Data were obtained through the IM construct with quartz reference on Ge-GDP films with initial density ρ_0 = 1.13 g/cm^3 (magenta circles, squares, and diamonds) and 1.1 g/cm^3 (purple square), where circles, diamonds, and squares represent 0.7%, 0.66%, and 0.5% Ge doping. Measurements are compared with LEOS models 5312 (dashed purple) and 5315 (solid orange), assuming 0.2% and 0.5% Ge doping, respectively.

FIG. 5. (Color online) Principal Hugoniot data and models for Ge-GDP in P–ρ plane, with Ge-GDP film initial densities and Ge doping as previously described. Data are compared with available Ge-GDP LEOS models, evaluated at an initial density ρ_0 = 1.13 g/cm^3. Models used to compare the GDP data are included as reference. Random uncertainties are shown as the colored (smaller) error bars and total uncertainties (quadrature of random and systematic uncertainties) as the gray (larger) error bars.

FIG. 6. (Color online) Ge-GDP results in the pressure–compression plane, with models and data as described in previous figures, showing structure observed in P–ρ plane is likely due to initial density variations.

FIG. 7. (Color online) GDP and Ge-GDP results in the pressure–compression plane show these ablator materials reach similar compression states with increasing pressure. The measured behavior follows similar trends to that predicted by LEOS models. Models and data as described previously.

FIG. 8. (Color online) Percent differences between quartz model predictions (from Refs. 8 and 18) in density and pressure, as a function of quartz shock velocity, are shown as the dashed blue and solid pink curves. Here a positive number means that a higher value is predicted using laser quartz fit in comparison to Z-machine fit. Circles and squares represent percent differences in density and pressure between IM results, assuming a reflected Hugoniot to calculate release states in quartz (orange, light blue, green, and purple circles and squares) and from inferred single-shock states using double-shock measurements (red circles and squares).

Tables

Table I. Material properties and associated errors at ambient conditions for ablator material films, as obtain for each batch. The shot numbers that used each of the batches are also indicated below.

Material	Formula	Ge (at %)	ρ_0 (g/cm^3)	n(λ = 532 nm)
GDP*	$CH_{1.36}O_{0.01}$	—	1.06±0.02	1.571±0.005
GDP†	$CH_{1.38}O_{0.02}$	—	1.05±0.05	1.571±0.005
GDP‡	$CH_{1.35}O_{0.01}$	—	1.044±0.005	1.563±0.010
Ge-GDP§	$CH_{1.42}O_{0.04}$	0.7	1.13±0.05	1.572±0.005
Ge-GDP¶	$CH_{1.43}O_{0.05}$	0.66	1.13±0.05	1.572±0.005
Ge-GDP‖	$CH_{1.40}O_{n/a}$	0.5	1.13±0.05	1.572±0.005
Ge-GDP**	$CH_{1.35}O_{0.01}$	0.5	1.10±0.05	1.570±0.010

*Shots 54126, 54127, 54185, 54187, 52635
†Shots 55774, 55775, 55777–55785
‡Shots 57162–57164
§Shots 54127, 54185, 54187, 52630
¶Shots 55774, 55775, 55777–55785
‖Shots 56115–56118
**Shots 57162–57164

Table II. Principal Hugoniot results for glow-discharge polymer (GDP) using the IM technique with quartz reference. Measured shock velocity with error is given for both quartz ($U_{s,Q}$) and GDP ($U_{s,GDP}$). The resulting particle velocity ($U_{p,GDP}$), pressure (P_{GDP}), and density (ρ_{GDP}) of shocked GDP are listed, with given random and systematic uncertainties. Random uncertainties enter the IM analysis through errors in shock-velocity measurements and initial density variations, while systematic errors stem from uncertainties in quartz's principal Hugoniot and release states.

Shot	$U_{s,Q}$ (μm/ns)	$U_{s,GDP}$ (μm/ns)	$U_{p,GDP}$ (ran,sys) (μm/ns)	P_{GDP} (ran,sys) (Mbar)	ρ_{GDP} (ran,sys) (g/cm^3)
55782	13.12±0.09	13.77±0.10	8.47(0.14,0.21)	1.22(0.02,0.03)	2.73(0.08,0.11)
55783	14.42±0.11	15.72±0.10	9.81(0.16,0.22)	1.62(0.03,0.03)	2.79(0.08,0.12)
55781	16.44±0.11	18.20±0.10	11.97(0.19,0.1)	2.29(0.04,0.02)	3.07(0.1,0.05)
55780	18.64±0.10	21.08±0.10	14.03(0.2,0.13)	3.11(0.04,0.03)	3.14(0.09,0.06)
55785	18.90±0.10	21.55±0.10	14.25(0.2,0.13)	3.22(0.05,0.03)	3.10(0.09,0.05)
55779	20.24±0.10	23.09±0.10	15.53(0.21,0.15)	3.77(0.05,0.04)	3.21(0.1,0.07)
55784	20.46±0.13	23.07±0.10	15.79(0.24,0.16)	3.82(0.06,0.04)	3.33(0.11,0.07)
55778	21.12±0.09	24.06±0.10	16.39(0.22,0.17)	4.14(0.06,0.04)	3.29(0.1,0.07)
55777	22.76±0.10	26.05±0.10	17.93(0.24,0.21)	4.90(0.07,0.06)	3.37(0.11,0.09)
52635	24.14±0.10	27.77±0.10	19.20(0.15,0.24)	5.65(0.05,0.07)	3.44(0.07,0.1)
55774	24.45±0.10	28.03±0.23	19.56(0.25,0.25)	5.76(0.08,0.07)	3.47(0.13,0.1)
55775	24.68±0.11	28.38±0.11	19.76(0.26,0.26)	5.89(0.08,0.08)	3.46(0.11,0.1)
54127	24.65±0.20	28.44±0.20	19.67(0.25,0.26)	5.93(0.08,0.08)	3.44(0.12,0.1)
54187	25.90±0.11	29.87±0.15	20.87(0.16,0.29)	6.61(0.06,0.09)	3.52(0.08,0.11)
57164	26.84±0.11	30.99±0.10	21.85(0.28,0.32)	7.07(0.09,0.1)	3.54(0.11,0.12)
54126	28.19±0.12	32.79±0.16	23.01(0.18,0.35)	8.00(0.07,0.12)	3.55(0.08,0.13)
54185	29.31±0.11	33.94±0.16	24.11(0.17,0.38)	8.67(0.07,0.14)	3.66(0.08,0.14)
57162	29.80±0.10	34.63±0.10	24.66(0.31,0.40)	8.91(0.11,0.14)	3.63(0.12,0.15)
57163	34.15±0.10	40.10±0.10	28.77(0.35,0.52)	12.04(0.15,0.22)	3.69(0.12,0.17)

Table III. Initial density and stoichiometry for available LEOS models for GDP and Ge-GDP.

Model	ρ_0 (g/cm^3)	Carbon (at %)	Hydrogen (at %)	Oxygen (at %)	Germanium (at %)
LEOS 5310	1.04	42.34	57.15	0.51	0
LEOS 5312	1.053	42.25	57.04	0.51	0.2
LEOS 5315	1.073	42.12	56.87	0.505	0.5

Table IV. Principal Hugoniot results for germanium-doped glow-discharge polymer (Ge-GDP) using the IM technique with quartz reference. Measured shock velocities in quartz ($U_{s,Q}$) and GDP ($U_{s,GeGDP}$) are given with measured error. The resulting particle velocity ($U_{p,GeGDP}$), pressure (P_{GeGDP}), and density (ρ_{GeGDP}) of shocked Ge-DP are listed with random and systematic uncertainties associated with each measurement. Random uncertainties enter the IM analysis through errors in shock-velocity measurements and initial density variations; systematic errors stem from uncertainties in quartz's principal Hugoniot and release states.

Shot	$U_{s,Q}$ (μm/ns)	$U_{s,GeGDP}$ (μm/ns)	$U_{p,GeGDP}$ (ran,sys) (μm/ns)	P_{GeGDP} (ran,sys) (Mbar)	ρ_{GeGDP} (ran,sys) (g/cm^3)
55782	12.98±0.11	13.56±0.09	8.20(0.15,0.21)	1.26(0.02,0.03)	2.86(0.09,0.11)
55783	14.50±0.09	15.21±0.11	9.83(0.14,0.22)	1.69(0.03,0.04)	3.20(0.1,0.14)
55781	15.97±0.10	17.28±0.12	11.37(0.17,0.11)	2.22(0.04,0.02)	3.3(0.11,0.06)
55780	18.64±0.10	20.76±0.11	13.83(0.19,0.12)	3.25(0.05,0.03)	3.38(0.1,0.06)
55785	18.84±0.10	20.87±0.27	14.04(0.19,0.12)	3.31(0.05,0.03)	3.45(0.15,0.06)
56118	19.24±0.11	21.18±0.09	14.44(0.2,0.13)	3.46(0.05,0.03)	3.55(0.11,0.07)
55779	20.29±0.10	22.5±0.11	15.41(0.2,0.15)	3.92(0.05,0.04)	3.59(0.11,0.08)
55784	20.44±0.12	22.81±0.10	15.53(0.22,0.15)	4.00(0.06,0.04)	3.54(0.11,0.07)
55778	20.91±0.10	23.53±0.19	15.94(0.21,0.16)	4.24(0.06,0.04)	3.5(0.12,0.07)
55777	22.66±0.10	25.53±0.16	17.59(0.23,0.2)	5.08(0.07,0.06)	3.64(0.12,0.09)
56117	23.37±0.09	26.53±0.11	18.23(0.23,0.21)	5.46(0.07,0.06)	3.61(0.11,0.09)
52630	24.18±0.10	27.35±0.10	19.02(0.24,0.24)	5.88(0.07,0.07)	3.71(0.11,0.10)
55774	24.34±0.10	27.87±0.11	19.11(0.24,0.24)	6.02(0.08,0.07)	3.59(0.11,0.10)
55775	24.5±0.12	27.76±0.12	19.31(0.25,0.24)	6.06(0.08,0.08)	3.71(0.12,0.11)
54127	24.7±0.10	28.02±0.18	19.49(0.24,0.25)	6.17(0.08,0.08)	3.71(0.13,0.11)
57164	27.04±0.10	31.14±0.11	21.78(0.27,0.31)	7.45(0.09,0.11)	3.65(0.11,0.12)
56116	26.82±0.10	30.75±0.20	21.45(0.26,0.30)	7.45(0.1,0.10)	3.73(0.12,0.12)
54185	29.22±0.09	33.72±0.09	23.67(0.28,0.36)	9.02(0.11,0.14)	3.79(0.11,0.14)
57162	29.90±0.10	34.57±0.15	24.47(0.29,0.39)	9.29(0.12,0.15)	3.76(0.12,0.15)
56115	31.80±0.10	36.65±0.10	26.12(0.31,0.44)	10.82(0.13,0.18)	3.93(0.12,0.16)
57163	33.89±0.10	39.64±0.14	28.20(0.33,0.50)	12.27(0.15,0.22)	3.80(0.12,0.17)