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LDRD PROJECT NUMBER: 215947
LDRD PROJECT TITLE: Coupled Experiment and Theory to Explore the Limits of Material Strength at High Strain Rates
PROJECT TEAM MEMBERS: Mitchell Wood(1444), James Stewart(1881) and Joseph Olles(2554)

ABSTRACT: (250 word limit)
Critical components such as detonators in Sandia's stockpile are heterogeneous materials that rely on accurate constitutive models for each material used in the component. However, experiments to study materials at high pressure are challenging and time consumptive, therefore we turn to modeling tools to refine and predict outcomes beforehand. Efficient models balance absolute physical accuracy against approximate but computationally lightweight constitutive inputs. By using a relatively small number of high fidelity simulations and experiments we have be able to broaden the predictive power of the shock response in metals and polymers. We have tailored the analysis of these simulations to determine a size dependent material strength, which can be used as constitutive model inputs for continuum hydrodynamics codes. For the shocked Cu, MD simulations show a yield strength from RMI jet growth of approximately 450MPa that depends on the details of the free surface geometry. This value is close to the yield strength of 500MPa parameterized for an elastic-perfectly-plastic strength model from experiments at the Dynamic Compression Sector at Argonne National Lab. The same analysis applied to MD simulations of PMMA jetting resulted in no clear determination of yield strength, implying a more complex RMI process in polymeric materials. Atomistic simulations of both materials are shown to be valuable training metrics and show the need for explicit strain rate dependent strength for future improvements in strength models used in continuum codes.

INTRODUCTION:
Measurements of material strength at high strain rates is of particular interest to the condensed matter and engineering science communities. Shock waves generated from gas guns, lasers or explosive drivers have been used to access strain rates ranging from $10^7 - 10^8 s^{-1}$, which provide ideal loading conditions to measure strength in a shock reflection setup. Planar shock impact has been used for strength measurements where the reflection within the material causes tensile loading at a spall plane, Hopkinson bar tests can be used to study lower pressure regimes where spall occurs and shear experiments have also been used for strength[1,2].

Complement to these experimental efforts, continuum hydrocodes have been extensively used to try and capture this high rate behavior in order to design explosive components and understand material failure in extreme conditions. As inputs these computational tools use parameterized constitutive models such as simplified Equations of State(EoS), thermal conductivities and most importantly, a strength model. The central approximation made in continuum hydrodynamics codes is not much of an approximation when very strong shocks are considered, as most materials can be well described as viscous fluids at that point. However, much attention has been placed on

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sub-critical or accident scenarios for components such as explosive foil initiators(EFI) used in the stockpile. While strong shock waves propagating in the EFI can be reliably predicted from continuum hydro codes, flyers that result from the EFI bursting bridge do not match well to empirical data. Modeling efforts without accurately parameterized material strength are the culprit of this shortcoming. Mismatches between experimental observations and model predictions underpins this work to look for new routes to calibrate strength models, especially for weak shock and accident conditions which are experimentally challenging to capture.

From a 1-D loading of a plate impactor, shock loading of a material with a non-planar free surface gives access to non-planar deformation, thereby exhibiting the presence of strength[3-7]. Ejected material from the free surface deforms in tension as it leaves the sample, and is considered as the limiting case of a Richtmeyer-Meshkov Instability (RMI). A RMI develops when a shock wave is transmitted through two materials that share a spatially non-uniform interface (nearly always the case), the amplitudes of this RMI are proportional to the Atwood number that compares the density of either material at the interface. For example, the Atwood number between a metal(material A) and a low density gas (material B) is given by $(\rho_A - \rho_B)/(\rho_A + \rho_B) \cong 1$. The surface perturbations at the interface can cause material to form 'jets' to form due to a convergence of compressive flow at these perturbations.

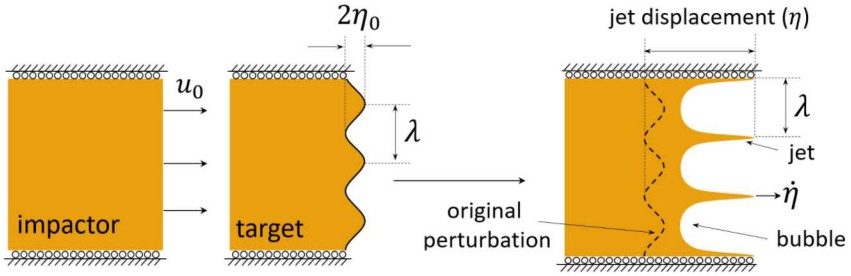

Figure 1: Schematic of impactor and target with perturbations. (Left) impactor has a defined velocity u_0 which drives a shock into the target having a set wavelength, λ, and amplitude, $2\eta_0$.

To experimentally probe material strength, specialized shock reflection experiments are conducted by members of this team at the Dynamic Compression Sector at Argonne National Lab. A corrugated surface is machined into metal and polymer plates so that shock waves that reflect off this surface jets material, this instability is imaged using synchrotron x-ray beams which characterize at sub-microsecond time scales. A schematic of these experiments is given in Figure 1, where jet displacement is used as the primary diagnostic to parameterize material strength. However, some experiments in this idealized geometry and loading conditions show anomalous jetting and bubble growth, leading to concerns of our understanding and modeling capability of strength at high strain rates[8].

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Observations from experiment will continue to inform and constrain the constitutive inputs to shock-hydro codes, there is risk of 'overfitting' to experiments which degrades model predictability. We are proposing to address if atomistic simulations can add to the training diversity in a way that builds more predictive engineering models. We would like this exploratory work to demonstrate 1) (Engineering outcome) that MD has validity in calibrating constitutive strength models for hydro code simulations and 2) (Scientific outcome) show there is/isn't an intrinsic size limitation for observed strength of a material in the prescribed shock geometry. We will discuss these two goals using two starkly different material systems, single crystal copper and polymer networks of Poly(methyl methacrylate) (PMMA). The former being a well-tested material standard, and the latter having very few conclusive experiments that have constrained the strength model parameterization. If successful, MD results of metal and polymeric spall will be used as training data for constitutive models in Sandia's hydro codes for both metals(Cu) and polymers(PMMA) of interest. If highly successful, follow on work will derive new strength model forms for polymers that is broadly useful for shock response. However, if this work fails to meet conclusive evidence on the aforementioned goals, the results will indicate that the anomalous deformation seen in experiments is due to microstructure features beyond the scale of MD. This is still valuable information when selecting and training strength models. Also, interpretation of experiments is easier if sub-μm deformation mechanisms can be ruled out.

DETAILED DESCRIPTION OF EXPERIMENT/METHOD:

To experimentally measure the RMI instability, photonic Doppler velocimetry (PDV) is used to capture the velocity of the jet and impactor [9,10]. Frequency-shifted PDV [11] was utilized in all experiments discussed here, where two fiber lasers are set slightly off-frequency from one another with wavelengths near 1550 nm. One laser was sent to the target and the other used as a reference. A single mode, collimated gradient-index lens probe having approximately 0.5 mm spot size, was used to send and receive the light passing through a circulator. Having a known time offset from one another, return and reference light were recombined to create a beat frequency in a fiber optic receiver, then digitized and recorded with an oscilloscope. The recorded beat frequency indicates the velocity of a reflective surface, here the impactor and jet.

To validate the velocity and gather the jet profile XPCI was used at Argonne's APS in the DCS [12,13]. Eight successive images were captured using a four-camera system of successive x-ray pulses from each shot. Images were acquired during 24-bunch mode, using a 1st harmonic undulator with white beam tuned to ~18 keV. The beam is operated in standard mode with one 80 ps pulse (33.5 ps FWHM) every 153.4 ns, and a slow shutter (60 ms) synchronized with the impactor (projectile) launch. The camera frames consisted of 1024x1024 pixel images, and having a pixel resolution of ~2.5 μm with the optical setup used[7,14]. The x-rays are manipulated to generate a spatially coherent beam where a propagation based, phase diffraction effect is observed on a scintillator. XPCI is a technique sensitive to refractive Features within the beam path are enhanced by contrast such as edges or voids, which are proportional to density.

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Regarding the atomistic simulation efforts, the most significant approximation made in any molecular dynamics(MD) simulation is in the choice of the interatomic potential(IAP). In contrast to higher fidelity electronic structure simulations, an IAP re-casts the full many-body interactions from electrons into a short ranged description of the atomic forces[15]. However, this allows for much larger and longer simulations due to favorable computational cost, which scales with the number of atoms, N, as $\sim N^1$ in MD versus $\sim N^3$ density functional theory[16]. Significant amount of effort is required to parameterize an IAP from scratch, and as such we have used well documented and properly tested IAP from the literature for this work.

The open-source Large-scale Atomic Molecular Massively Parallel Simulator (LAMMPS) atomistic code is utilized to perform a series of classical molecular dynamics (MD) simulations to predict the yield strength of Cu via the Richtmyer-Meshkov instability (RMI) [17]. The embedded-atom method (EAM) interatomic potential, parameterized for Cu by Mishin *et al.* [18], is employed to describe all interatomic interactions within the single-crystal Cu samples. This potential is chosen as it has been shown to capture experimentally observed global defect dynamics and microscopic lattice responses of shocked Cu in very large-scale MD simulations [19, 20].

A set of pseudo-two dimensional (2D) single-crystal Cu structures are created within LAMMPS that contain a single sinusoidal perturbation located at the +Z surface with a wavelength of $\lambda = 2\pi/k$, where k is the wave number. The size of the simulation domain in the Y-direction is taken to be ~3.5 nm while the X-direction contains the surface perturbation (i.e., the length of the X-dimension is the wavelength). The length of the Z-direction is taken to be 5 times the wavelength to delay any self-interaction associated with the reflecting shock wave. Three crystallographic directions, [001], [011], and [111], oriented along the Z-axis, are chosen to study the yield strength of Cu using the RMI. Wavelengths of 10 nm (~155,000 atoms), 50 nm (~3,700,000 atoms), 100 nm (~15,000,000 atoms), and 500 nm (~380,000,000 atoms), which are readily accessible by MD, are chosen to study the size effects in calculating the yield strength from the RMI method. Furthermore, three non-dimensional wave numbers are also chosen to study the size dependent jetting behavior of Cu, $\eta k = 0.5, 1.0, 2.0$. Note, at the continuum scale, $\eta k < 1$ predicts growth and arrest behavior while $\eta k \gtrsim 1$ predicts growth and breakup behavior.

All structures are initially equilibrated to 300 K for 100,000 timesteps (100 ps). Periodic boundary conditions are used in the X and Y dimensions to simulate bulk-like samples. Shock is induced in the Z-direction using an effective infinite-mass piston to drive the system with a defined velocity. The piston velocities, U_P, studied in this work include 0.75, 1.00, 1.25, 1.50, 1.75, 2.00, 2.25, and 2.50 km/s. All simulations are performed using the microcanonical or NVE ensemble (i.e., fixed number of atoms, volume, and total energy) with a 1 fs time step.

During the course of the shock simulations, average atom positions at the perturbation peaks (i.e., $kx = 0, 2\pi$) and perturbation minimum (i.e., $kx = \pi$) are tracked in order to determine (i) the distances the free surfaces and jet traveled, (ii) the jet length, and (iii) the instantaneous jet and free-surface velocities. With this information, the yield strength can be calculated via the following expression [3, 9]

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$$Y_{RMI} = 0.24\rho_{Cu}\frac{\left|V_{Jet}^{0}\right|^{2}}{kh_{Jet}^{max}}$$

where ρ_{Cu} is the ambient density of copper (8.96 g/cm^3), $k = 2\pi/\lambda$ is the wave number, h_{Jet}^{max} is the maximum jet amplitude at the moment of saturation as measured from the initial free-surface position, and V_{Jet}^{0} is the peak jet growth-rate defined as the difference between the maximum jet velocity and the corresponding free-surface velocity.

For the polymeric system studied here, there are plenty of available IAP in the literature, with a wide range of computational cost associated with them. At the most expensive end there are 'fully-reactive' potentials such as ReaxFF[21] which describe chemical bonds for every atom through a set of bond orders that are dynamically calculated. This means that bonds can break and reform as a simulation progresses, in contrast to cheaper IAP that assign bonds as initial conditions and will not break due to a harmonic potential between bonded atoms. As an exploratory effort, we used a non-reactive polymeric IAP in OPLS[22], which also omits partial atomic charges in order to avoid expensive long range electrostatic solvers in MD[23]. This potential has been used in similar simple polymers[24], and has been shown to agree with experimental values of bulk moduli and glass transition temperature[25].

Unlike solids with a defined crystal structure, building a representative input structure of a polymer network for use in an all-atom MD simulation is non-trivial. As such there are a number of specialty software programs such as PolymerModeler[26], MaterialsStudio[27] and Enhanced Monte Carlo[28] that all take a unique approach to 'grow' polymers in a simulation cell. For example, EMC uses a Monte Carlo step to determine the conformation of each repeat unit as it is added, followed by a short annealing step to relax the overall structure. Samples prepared for this study used EMC to build a semi-periodic slab of Poly(methyl methacrylate) (PMMA) with an aspect ratio of 5:1:1 where the longest direction (shock direction) is non-periodic which prohibits polymer chains to connect through the boundary. Molecular weight(M_w) of polymer chains will have a significant effect on both mechanical and thermal properties[29,30], as such we chose to contrast the shock response of very short ($M_w = 10$ mer/chain) and long ($M_w = 200$ mer/chain) polymer networks. Each structure was built at the experimental density of $1.18 g/cm^3$ with a Monte Carlo and relaxation temperature above the glass transition($T = 1000K$) and chains were terminated with a methyl group. While PMMA slabs of variable size were generated, only the largest will be discussed here which contains ~10M atoms and takes several days of compute time to construct the initial structure.

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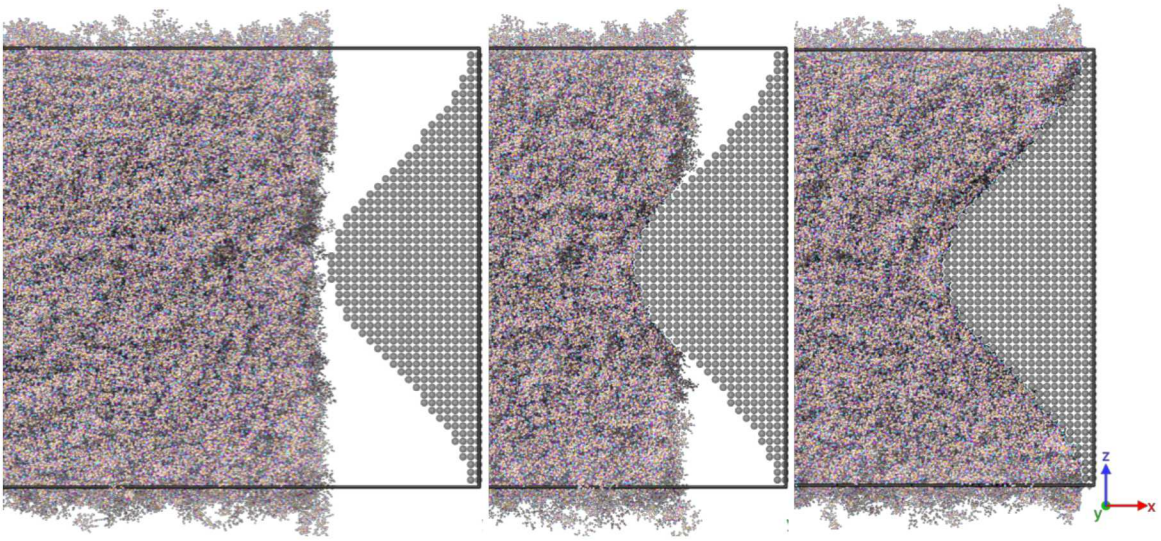

Figure 2 Time series of the surface geometry preparation for PMMA within MD. The shaped indenter (grey spheres) is moved at a constant velocity to the left as the PMMA is held above its' glass transition temperature. Cooling through the glass transition with the indenter present preserves the shape after it is removed.

Prior to the intended shock simulations, the following relaxation procedure was used to prepare the PMMA structures. All MD simulations were performed using the Sandia developed LAMMPS software package[17]. As built structures were held at the build temperature (T=1000K) for 1ns before being cooled to 300K over 1ns and then subsequently held at 300K for an additional 1ns. Each of these thermalization steps used a Langevin thermostat[31]maintaining fixed cell lengths in the periodic directions (Y,Z) while the non-periodic (X, eventual shock direction) is allowed to vary. Next the system is allowed to relax in an isobaric state with the pressure barostat set to 1 atmosphere, this causes the aspect ratio of the slab to change. To replicate the surface geometry used in the APS experiments, a solid intender was constructed of carbon atoms to create the $\eta_0 k$ values of 0.5, 1.0 and 2.0. To retain this surface structure, the polymer was heated to 600K (above the glass transition for PMMA) while the indenter was pressed into the surface and then cooled to 300K to imprint the polymer surface. An example time series of this process is given in Figure 2, the carbon atoms of the indenter (grey spheres) have their forces zeroed at each timestep meaning its' shape does not change through the simulation. The final dimensions of the $M_w = 200$ system was $276x28x28nm$ and $389x24x24nm$ for the $M_w = 10$ PMMA slab.

Similar to the Cu MD work, a shock wave is generated in these polymer systems by placing an infinitely massive (momentum mirror) piston at one end of the sample and all atoms are assigned a velocity toward the piston, this leaves shocked material stationary in the laboratory reference frame. Analysis of temperature, stress, density and velocities are locally averaged on a $1nm^2$ grid.

RESULTS:

One of the striking results from the APS shots to date was the enormous differences in jetting behavior the PMMA samples showed from any of the metals tested prior. Even at very high shock

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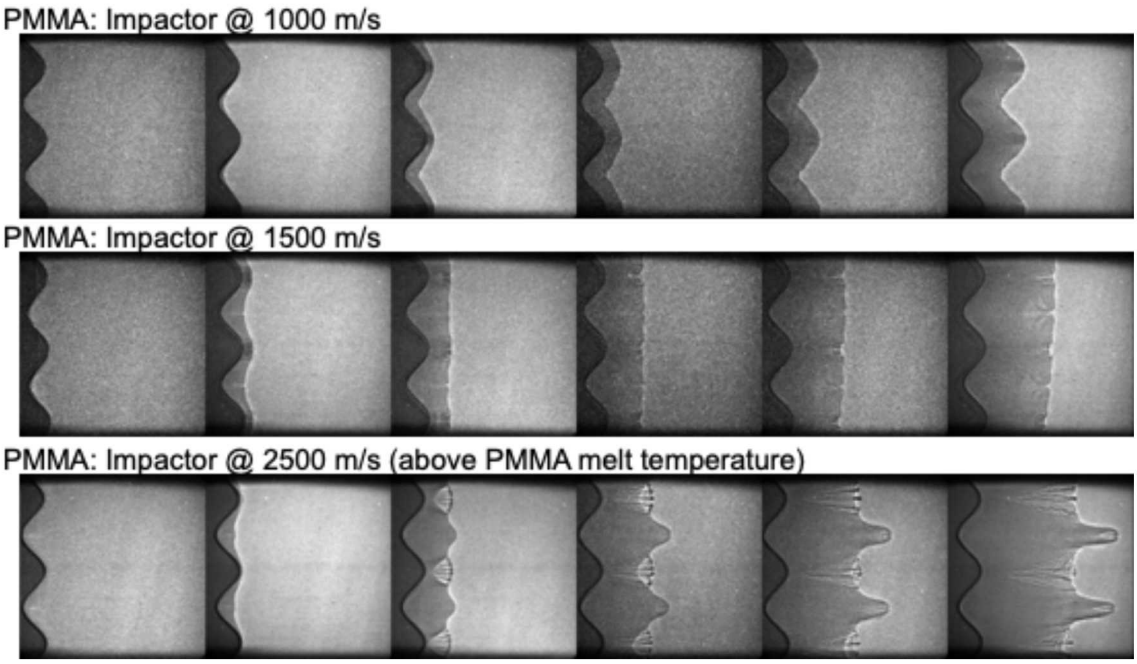

Figure 3 (Top) Image sequence of a PMMA target with $\eta_0 k = 1$ and $\lambda = 200\mu m$ impacted at 1km/s, each frame is separated by 153.4ns, this would be classified as 'No Growth' (Middle) Same geometry but impacted at 1.5km/s, this is a borderline NG or G&A and would depend on the details of the velocity profiles. (Bottom) Time series of the same geometry impacted at 2.5km/s, resulting in a temperature increase above melt. It is unclear if the polymer jet will arrest or breakup at later times.

pressures, where one could assume 'fluid-like' flow, it is hard to explain the non-uniform density profiles at the leading edge of the jet and in the spall zones(See Figure 3). To gain better insight into this process, MD simulations were run spanning a large range of shock strengths (U_p = 0.50 to 3.00km/s) for three different surface wavenumbers (0.5, 1.0 and 2.0). In addition, it was our hypothesis that the molecular weight of the polymer chains would act analogous to the critical feature length that was the focus of the Cu MD work. As an exploratory effort we aim to bracket the extremes of this behavior with a very low molecular weight structure ($M_w = 10$) and one that is high by MD standards($M_w = 200$). Although the long chain case still seems low by experimental measurements[32], MD work using the same force field on simple linear polymers shows a convergence of the thermal conductivity around a molecular weight of a few hundred, implying bulk-like behavior at these chain lengths[33]. Conversely, the low molecular weight PMMA will have limited chain entanglement and will behave more like a wax than a glassy polymer like the longer chain sample.

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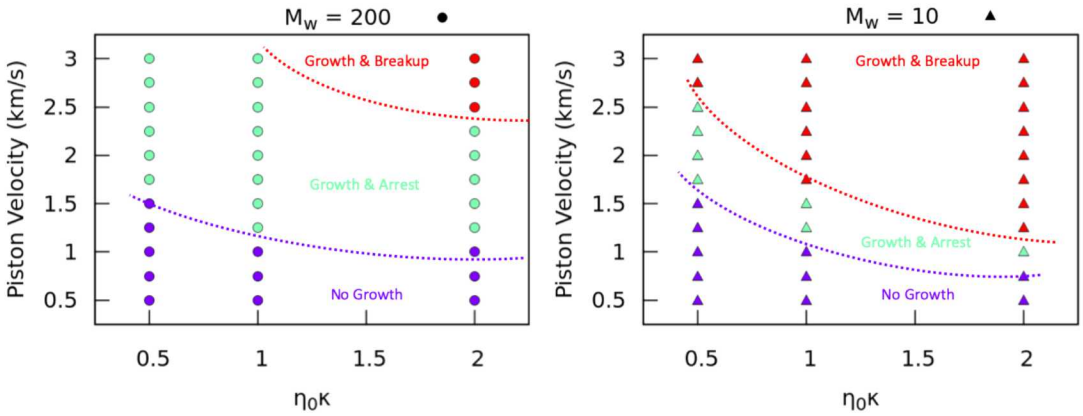

Figure 4 (Left) Jetting behavior for the high molecular weight PMMA samples. Only the strongest shocks with $\eta_0 k = 2.0$ were seen to eject polymer chains from the surface. (Right) Characteristic jetting behavior for the short chain PMMA samples, there is a noticeably smaller range of growth and arrest for these weakly entangled systems.

The collection of all PMMA shock simulations is displayed in Figure 4 and is classified using the language of Buttler et. al.[34] based on the behavior of the jetted (or lack thereof) material. To be classified as 'No Growth'(NG) the surface could maximally become planar during the shock reflection before returning to a structure that is self-similar to the original one. In addition, the free surface velocity of the NG cases could not saturate at long times equal to, or above twice the piston velocity in order to meet this classification. A classification of 'Growth and Arrest'(G&A) was given to samples that minimally became planar (but could invert) and remained so at late times. Free surface velocities here had to be equal to twice the piston velocity at late times in order to ensure there was no jet growth. Lastly, the designation 'Growth and Breakup'(G&B) was given to samples that minimally inverted the original surface profile and exceeded twice the piston velocity in the free surface velocity. In many cases polymer chains were ejected into vacuum leaving no restoring force to inhibit further jet growth. Dashed lines are superimposed on the data to imply

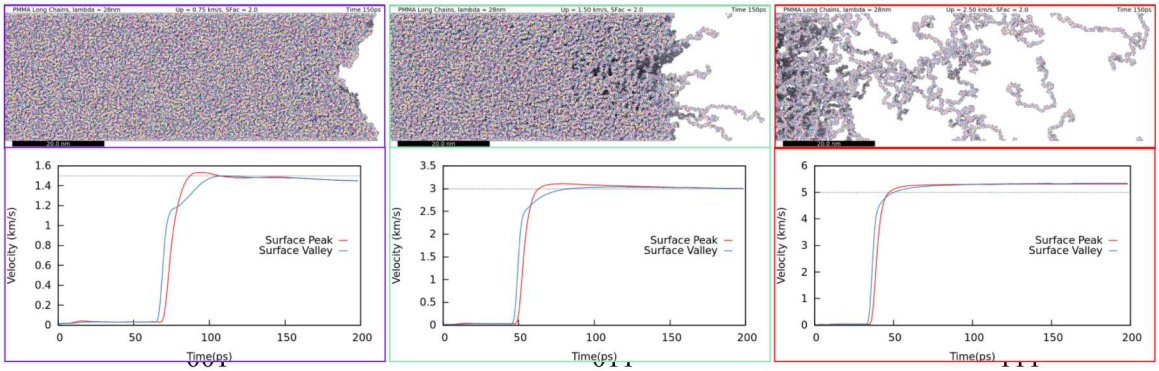

Figure 5 (Left)Top panel shows the shocked long chain PMMA simulation after 150ps where U_p=0.75km/s while the bottom panel tracks the surface velocities (5nm depth) of material originally at either the valley or peaks, see Figures 1,2. The horizontal dashed line is equal to twice the piston velocity. (Center) Example of growth and arrest, surface material travels together at twice the piston velocity. (Right) Example of growth and breakup for PMMA where whole chains are ejected from the surface, surface velocities are seen in excess of twice the piston velocity.

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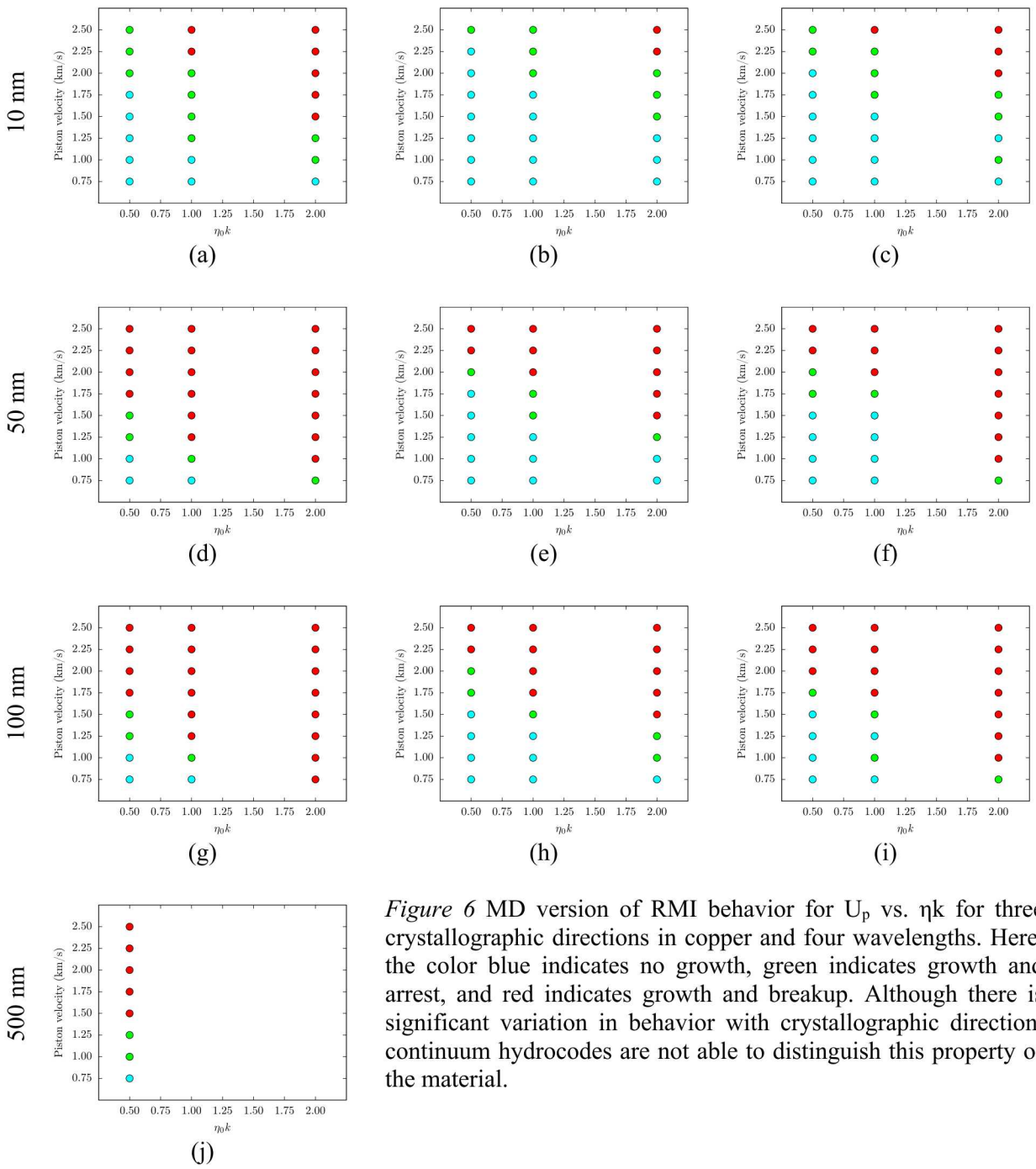

Figure 6 MD version of RMI behavior for U_p vs. ηk for three crystallographic directions in copper and four wavelengths. Here, the color blue indicates no growth, green indicates growth and arrest, and red indicates growth and breakup. Although there is significant variation in behavior with crystallographic direction, continuum hydrocodes are not able to distinguish this property of the material.

the boundary between these characteristic jetting behaviors if a continuous sampling of $\eta_0 k$ or U_p was undertaken.

Complementary data to the classifications made in Figure 4 are on display in Figure 5 where simulation images at late times and free surface velocity profiles are given for each classification.

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The data in Figure 5 are for the longer PMMA chains and $\eta_0 k = 2.0$ at piston velocities of 0.75km/s (left), 1.50km/s (center) and 2.50km/s (right). The distinction of 'peak' and 'valley' in the lower panels of Figure 5 is in reference to the original positions of the material prior to the shock drive. The center of mass of a 5nm by 5nm chunk of material in the X-Z plane (see Figure 2 for direction assignment) is calculated using the post-processing and visualization software OVITO[35]. In each of these velocity profiles it is clear to see the shock arrives at the valley first, accelerating this material above the imposed piston velocity due to momentum conservation of the reflected release wave. Shortly thereafter the material at the peak of the surface profile is accelerated, and the final velocities of the surface material depends on the details of the jetting.

The full collection of the RMI characterization from MD on shocked copper is displayed in Figure 6, broken out by crystallographic direction and size of the surface perturbation. The largest set of simulations, with a 500nm wavelength, only display a partial set of data due to the high computational cost to generate this data. In each of these plots, red points represent G&B, green to G&A and blue for NG, using the same criterion as the PMMA shocks. For each size and orientation, there are shocks that result in each of these RMI characterizations which is encouraging to see that the EAM potential used here can capture the full range of behavior. Of all of these cases, those that are designated G&A are used to calculate the yield strength(Y_{RMI}) defined in the previous section. This data for Y_{RMI} is captured in Figure 7, where points of the same wave number and perturbation length have their values averaged across different piston velocities. An additional point is added on this plot for the yield strength used in the EPP continuum strength model, this value is constant across all perturbation lengths.

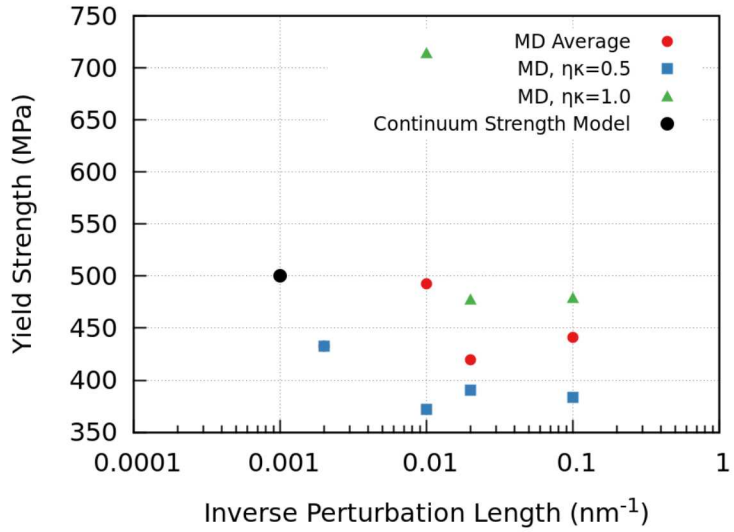

Figure 7 Calculated yield strength from MD simulations versus the inverse of the wavelength for copper along the [001] crystallographic direction for various piston velocities. The yield strength at each perturbation length are averaged of piston velocities that are classified as G&A.

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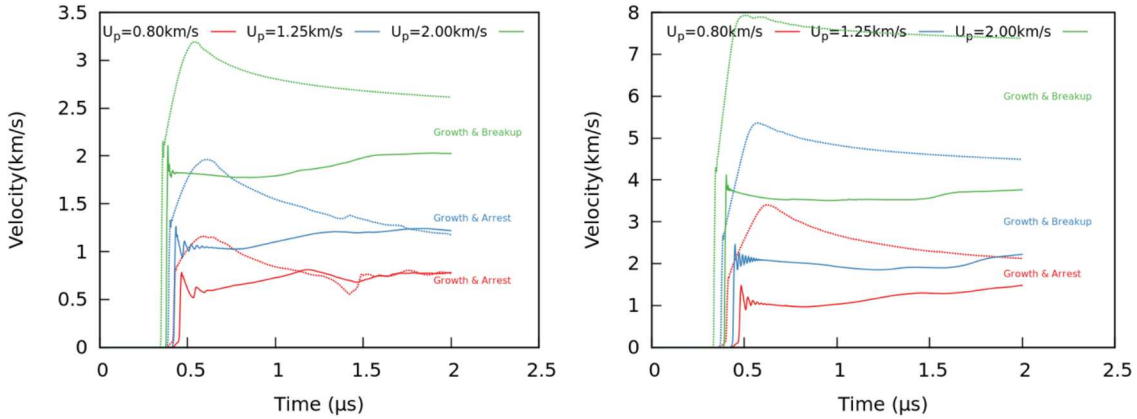

Figure 8 Velocity profiles from the 1mm wavelength ALEGRA Cu simulations using an EPP strength model with a yield strength set to be 500 MPa. (Left) $\eta_0 k = 0.5$ (Right) $\eta_0 k = 1.0$ The dashed lines are the jet (spike) and the solid lines are the bubble (free surface) velocities. If the pair of velocity profiles for each piston velocity asymptote to the same value, they are considered as 'Growth & Arrest' otherwise as 'Growth & Breakup'.

Lastly, A set of continuum simulations were conducted in order to draw direct comparisons to the MD shock simulations on Cu. Equivalent PMMA continuum simulations were not undertaken due to an inability to capture experimentally observed jetting behavior with available strength models in ALEGRA. In all of the continuum simulations conducted here, we were only able to observe G&A and G&B RMI behavior. Figure 8 displays the result of a few of these continuum simulations where the transition between G&A and G&B is observed. Moreover, there was no size dependence on the RMI from these continuum simulations, the velocity profiles of both jets and bubbles are exactly the same at 1μm as they are at 1mm, see Figure 9. This is expected since the EPP strength

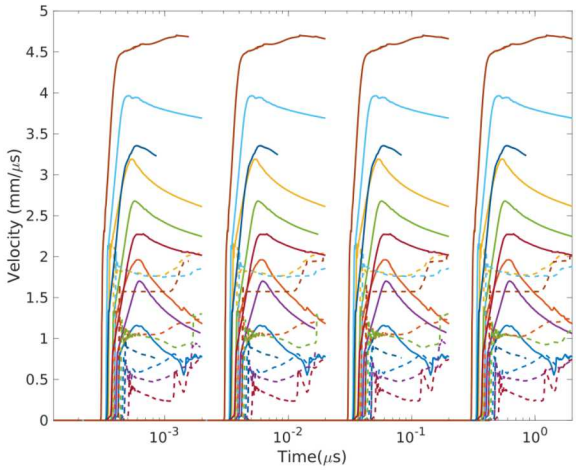

Figure 9 Velocity of the jet(solid) and bubble(dashed), with wavelengths 1, 10, 100, and 1000 microns from left to right, respectively. Wavenumbers vary from 0.5, 1.0, and 2.0, and within each wavenumber the impact velocity was varied from 0.8, 1.25, and 2.0 km/s. Note there is no change in velocity profiles with change in wavelength.

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model has no time dependence, and therefore any means to distinguish the deformation of material with 1μm or 1mm feature sizes.

DISCUSSION:

Through this work we aim to answer one key scientific question and one to address a more applied, or engineering outcome of the simulation work. The former is to determine a critical length scale at which non-ideal(i.e. viscoplastic) jetting and spall behavior gives way to hydrodynamic behavior where a shocked material is best represented as a viscous fluid. Holding the surface geometry and piston velocity fixed, this should be determined in the Cu samples by simply scaling the physical size of the simulation. However, the PMMA shock simulations carry a second characteristic length-scale which is the average length of the individual polymer chains. The latter project outcome is to determine the viability of MD simulations mimicking the shock experiments to be used as training data for constitutive models used in shock hydrodynamic codes such as ALEGRA. Previously this was done using only experimental data which limits the total number of training points since the experiments rely on specialty equipment with limited availability at Argonne National Lab. This second goal is important for both of our computational and experimental customers because it would demonstrate a viable supplement to these experiments that would reduce the empiricism in tuning inputs into important engineering design/decision codes. Both of these outcomes will be discussed further here, for each of the materials studied.

Starting with the results of the Cu MD shock simulations, looking at Figure 6 there are clear changes in RMI character based on the size of the perturbation, notably there are a larger proportion of U_p and $\eta_0 k$ that result in G&B when moving from the 10nm to 50nm perturbations for [001] directed shocks. Furthermore, there seems to be a strong orientation dependence for the smallest perturbations (10nm), but this difference is diminished as the perturbations reach 100nm in size. From this data alone one may assume a 'critical' length scale on the order of tens of nanometers, but is not clearly corroborated by the Y_{RMI} data in Figure 7. There is significant scatter in the MD calculated strength for λ<100nm, and we were only able to capture G&A in one of the 500nm sized simulations. These calculations are quite sensitive to the measurement of h_{Jet}^{max} which may not be fully resolved given the limited timescale of MD simulations. That said, the measured Y_{RMI} from MD are not too dissimilar to the Cu yield strength used in the EPP model that was tuned to capture the jet behavior of actual RMI experiments. In fact, the $\eta_0 k$=1 data, where G&A is expected from continuum level descriptions, show a calculated Y_{RMI} the closest to the experimentally tuned EPP model. Regarding the first of this project's goals, it is undetermined if we have measured a critical length scale for hydrodynamic jetting due to uncertainties in the measurement of yield strength at MD length/time scales. Given time to refine the measurement and also complete our dataset for λ=500nm would provide a clearer answer. Regarding the second of this project's goals, there is a clear advantage in using MD as training data for continuum strength models. The only strength model whose fit of the experimental measurement of jet velocity is properly constrained (i.e. number of free variables less than observables) is EPP, but this clearly just a qualitative representation of a shocked material since there is no strain rate(and thus size) dependence on jet behavior. While the MD data underpredicts the fitted EPP yield

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strength, there a far richer dataset of RMI (or lack thereof) from MD than what was able to be gathered from a reasonable amount of time at APS-DCS. In addition, there exists local temperature, stress, strain and strain rate data from MD that we were not able to analyze in detail here that can be used to train strength models.

As for the polymeric shocks, given the complex nature of the setup and relaxation steps needed in order to properly conduct the PMMA MD simulations, only a limited number of domain size and chain lengths were studied here. However, to fully characterize the shock response a wide range of shock strengths and surface wavenumbers were studied here. We were able to clearly identify three different characteristic jetting behaviors in PMMA based on final jet morphology, ejected mass fraction and free surface velocities, this data is presented in Figures 4 and 5. Comparing the high and low molecular weight PMMA simulations, two conclusions can be drawn. First, the transition between the NG and G&A jetting characteristics is seen to be relatively insensitive to the molecular weight as well as changes in $\eta_0 k$. At a piston velocity of 1.25km/s where this transition is observed in both samples, a uniaxial pressure of 5.68GPa and a temperature rise to 540K is observed, the rarefaction wave off the free surface cools the sample down to 473K. From the MD results the primary factor in this qualitative change is the piston velocity, which actually agrees quite well with the experimental results, as can be seen in Figure 2. A $\eta_0 k$ value of 2 is used for all shots in Figure 5, where using the rules defined to characterize the jetting behavior(if available) the Top and Middle sets of images would be designated as NG and G&A, respectively.

The second conclusion of this work is in regards to the late time behavior of strongly shocked samples. A noticeable difference between high and low molecular weight systems is the suppression of the G&B jetting at higher molecular weights. This makes conceptual sense due to chain entanglements restricting the flow of the jetted material as well as the omission of bond breaking in these MD simulations. Though even at high rate deformation, one would expect significant crazing and chain pullout prior to covalent bond breaking given the vastly different energy barriers to inter- vs intra-molecular bond strengths. Extending this trend to much high molecular weight polymers used in experiments, there may be a complete loss of the G&B behavior. Looking at the highest piston velocity shown from experiments (2.50km/s, Figure 2 bottom panel), the surface has inverted but it is unknown what the relative velocities of the jet and spall zones are. Our MD predictions of $\eta_0 k = 1$, $M_w = 200$ and the same piston velocity were characterized as G&A.

Applying the same protocol for calculating $\mathrm{Y_{RMI}}$ as was done for Cu MD simulations results in largely unsatisfactory results for PMMA, where nearly all calculated yield strengths are $\approx 10^2 Pa$ which is far below the expected $\approx 60\mathrm{M}Pa$ measured from traditional tensile tests. Alternative routes for calculating $\mathrm{Y_{RMI}}$are needed given orders of magnitude disagreement between MD and experiment. This is somewhat expected since the $\mathrm{Y_{RMI}}$ measurement relies on jetted material velocities far exceeding free surface velocities, which was not observed here from MD or experiments.

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Given the visual agreement between the MD and experimental jetting behavior, it is reasonable to assume that the MD results could be used to calibrate strength models, in the same way that experiments have already provided. While it is a much larger challenge (than this effort has scope for) to determine if metal plasticity models like JC, SGL, etc. can be applied accurately to polymers in a shock hydro code, these strength models can at least be trained to reproduce the qualitative jetting behavior map that MD has provided for PMMA. Furthermore, the detailed information(sub-ns time resolved) of the free surface velocity profiles can be used to fit a strength model in the same fashion that the experimental jet displacement distances(~100ns time resolved) were used by Hudspeth et. al.[36]. The main criticism of this assertion to use MD as training data is whether or not the experiments quantitatively agree with the MD predictions of PMMA strength. Up to now the comparisons between MD and experiment have been drawn based images of either system during jetting. Assessing other material properties from MD that can be experimentally verified such as structure factors, thermal conductivity, or Hugoniot state points to name a few. The level of agreement needed in these quantitative measures would be at the discretion of the ALEGRA developer/user when training the new strength model. At the very least, the inclusion of MD data in training prevents the obvious problems of overfitting a strength model to the small experimental dataset.

The more basic science goal of this work regarding the critical length scale for ideal jetting in PMMA is best answered with what is missing from the current work to draw a clear conclusion. We would first have to build structures that have a larger cross section perpendicular to the shock direction, as was done for Cu. This will allow for a more detailed insight into the jetting behavior as a function of the physical size of the system. In these larger systems we hope to see material flow only from the valleys of the surface perturbation, allowing additionally for a detailed look at the spall zones in shocked PMMA which show interesting structure from experiments(See Figure 2 Bottom panel). As was mentioned previously, the second length scale in a polymer is in the molecular weight which we already noticed a characteristic change in the strong shock behavior between the two chain lengths tested here. It would be valuable to build samples at longer chain lengths closer to experiments in order to test if there are conditions where G&B still exists, or if this completely suppressed as this internal length scale is increased.

ANTICIPATED OUTCOMES AND IMPACTS:

There are several avenues for continued effort on this project theme of ‘multiscale calibration and validation of engineering codes’. This work has shown the abundance of detail available from MD simulations of shocks, and highlighted places were connections and validations to experiments can be clearly made. In doing so we now have a method to reduce the empiricism in long-standing engineering models that are currently in use at Sandia. The scientific impact of this work is deferred until we can complete our datasets on Cu and PMMA, as well as refine the measurements of strength calculation for the polymeric systems. We do expect at least one high impact journal publication from this work as these MD simulations demonstrate usage of cutting edge high-performance computing resources in a way that is novel to the shock physics community.

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Using MD at trivial and extreme length scales to elucidate the behavior of complex physical phenomena will lead to a transformative scientific and engineering understanding of shock physics and solid mechanics. Utilizing MD at these extreme length scales provides a direct bridge to fill the gap between modeling with atomic resolution and continuum level codes, such as CTH or ALEGRA, that are widely used to impact Sandia's missions. Furthermore, such extreme computing drives the innovation of state-of-the-art computing resources and accelerated modeling techniques. As MD does not explicitly incorporate any continuum equation-of-state (EOS) to govern a system's dynamics, MD allows for the generation of large datasets to interrogate fundamental and coupled physical phenomena, thus allowing for the discovery, development, and validation of predictive models, in both normal and harsh environments, that are ultimately incorporated into component and systems level models.

We believe there are several funding calls internal to Sandia that this work is well posed to meet. First, the most recent calls for full LDRD projects in FY20 contained language that this work would fall under. Specifically, within the ND mission campaign call the subject area entitled *'Development Agility: Integrated Qualification'* has a central goal of accelerating the qualification process of the NW development cycle. Growing upon the effort reported here, we would be able to specifically address the *'ModSim/CompSim data used as qualification data'* and *'Design simulation data used as qualification data'* topics of this unit of this ND call. We see an opportunity to integrate MD/continuum coupled simulations to provide supplement to experimental qualification of components. Given the nature of computational efforts, numerous tests of standard operation and edge cases can be evaluated in a predictive manner prior in order to plan an optimal design of qualification experiments. Since the calibration of material strength models is often the slowest (depends on results of experiments) and most empirically tuned(phenomenological versus predictive), accelerating this development would net an overall decrease in time-to-qualification of components used in the stockpile. The same argument can be applied to the language used in the LDRD ES research foundation FY20 call. A core topic of this call is *'Fundamental and Coupled Physics Phenomena'* wherein specific language of *'Discovery and development of predictive models for engineering science phenomena needed to predict the performance of components and systems. This includes models that capture environmental responses in both normal and harsh environments'*. We see this work of probing material strength of extremes of strain rate as an ideal match to the need for predictive simulation in extreme environments.

Parallel efforts to what we have developed here and what we are proposing as future work include Sandia's involvement in the TriLab (LANL, LLNL and SNL) consortium on material strength. The focus of this work, which is funded by the Science Programs Campaign 2, is to use an ideal material for shock response (tantalum) in order to make direct comparisons between continuum hydrocodes developed at each lab (i.e. ALEGRA, ALE3D) for experimental conditions across a wide range of pressures and loading paths (i.e. NIF, Z-Pinch, Explosive drivers). Speaking with a member of this team, Matt Lane (SNL, 1864), he summarized their work locally as understanding the role of microstructure on material strength, where their efforts are similarly attempting the bridge atomistic and continuum methods. The existence of such a large project, but being limited

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to just a material standard up to this point highlight the need for branching efforts like the one proposed here to make deliberate progress on 'Sandia Relevant' materials such as metals and polymers that are present in critical components such as detonators.

CONCLUSION:

From this work we have detailed the shock response and RMI characteristics of a pair of materials (Cu and PMMA) that are currently used in the stockpile, specifically in EFI. Understanding material strength at high rates of deformation is critical in making accurate predictions of the performance of these EFI detonators, and significant experimental effort has been invested to determine these effects. We have shown in this work that high fidelity atomistic simulations can be used to characterize material strength which can in turn be included into continuum hydrocodes that are used for component design and testing. To date, there have been very limited demonstrations of physics codes such as molecular dynamics informing constitutive model inputs used in engineering codes. Rather, continuum inputs are usually empirically tuned until sufficient agreement with experiments is achieved. Our atomistic/continuum/experimental comparison shown here on Cu details how a multiscale coupling can be done to fill the gaps left by experiments. For PMMA we have demonstrated qualitative agreement between MD and experiments where continuum codes are poorly posed to capture the same shock response and RMI of these polymers. Extensions of this work will truly enable predictive continuum simulations that are parameterized using information from both experiments and simulations on a wider class of materials than ideal metals and simple polymers.

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