44.0 ADVANCED CHARACTERIZATION OF PARTICULATE MATERIALS SIMULATING HIGH EXPLOSIVES

Summer Camerlo (Mines)
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This project initiated in Fall 2020 and is supported by the Center for Micromorphic Multiphysics Porous and Particulate Materials Simulations with Exascale Computing Workflows, U.S. Department of Energy (DOE), National Nuclear Security Agency (NNSA), Office of Advanced Simulation and Computing (ASC), ASC Predictive Science Academic Alliance Program III (PSAAP III). It involves collaboration with the University of Colorado Boulder and other academic partners, in addition to the three NNSA national laboratories: Los Alamos National Laboratory (LANL), Sandia National Laboratories (SNL), and Lawrence Livermore National Laboratory (LLNL). The experimental work performed during this project will serve as the basis for a Master's thesis program for Summer Camerlo.

44.1 Project Overview and Industrial Relevance

Traditional plastic bonded explosives or high explosives (HEs) require extensive safety precautions in handling and testing environments. HEs are of great interest to the NNSA, as well as other U.S. Department of Defense labs. Current knowledge of HEs is limited, from processing to thermo-mechanical behavior states for both virgin and recycled HEs. Thus, there is clearly a need for robust characterization of HEs, which can be used to inform predictive modeling of HE behaviors [44.1].

By using materials that emulate characteristics of traditional HEs for simulations and other non-critical testing, a considerable amount of time and money can be saved. This project is intended to provide a robust characterization of a mock high explosive (MHE) developed by LANL that contains angular idoxuridine (IDOX) crystals suspended in a binder matrix. Features like particle sizes, shapes, and distributions are of interest, in addition to the processing of these materials and their quasi-static to dynamic mechanical response. In this project, experiments will be performed to link MHE characteristics to processing and properties, building a database of information that will be used to inform multiscale computational efforts underway at the University of Colorado Boulder. Due to the highly sensitive and costly nature of experimental studies with HEs and MHE's, the availability of predictive computational models will enable parametric studies of these materials under a broader range of conditions, potentially including under different strain rates, shock loading, cyclic loading, and varied thermal conditions [44.2]. The end result will be a comprehensive database of in-situ/ex-situ quasi-static and high strain-rate compression testing data, with x-ray radiography and computed tomography (CT) whenever possible to fully characterize the particle/matrix characteristics. In this project, we will simulate with quantified uncertainty the deformation response of MHEs, which can then be modified to represent full plastic bonded explosives of interest to the NNSA labs.

44.2 Previous Work

The first objective of this project was to develop an experimental and computational framework to be used to study IDOX. Unfortunately, though IDOX is easier and safer to work with than HMX, it presents its own set of challenges. Its main component, iodine, has a high z number and it is difficult to get detailed CT images [44.3]. Additionally, its irregular crystalline structure and wide array of particle sizing, ranging from 10-600 microns, add difficulty for modeling. Thus, a subsection of the project became to create "surrogate" mocks for calibration of experimental systems and to build a robust modeling framework. For this, baseline data was initially obtained by experiments and modeling of epoxy embedded with particles. The "model" particles were glass beads of varying sizes, representing simple spherical particles embedded in the epoxy matrix, as well as fine, angular sand, which better represents the IDOX crystals in size and shape found in the LANL MHE. Ultimately, it was determined that glass beads in epoxy were too far removed from the reference material of IDOX in a polymeric binder and thus a new surrogate sample was chosen: F50 sand in the polymeric binder Kel-F.

Using specifications received from LANL, a powder compaction die was procured for creation of these new samples. To produce them, the die is heated to 50°C on the platen of the Genesis hydraulic press. A mixture of F50

sand and Kel-F binder is added and allowed to come to temperature. It is then compacted at two tons and broken out of the die at temperature. These surrogate samples were used for mechanical testing and CT modeling.

Additionally, LANL provided the project with machining fines recovered from pristine MHE production to see if "recycled" samples are viable options for testing. IDOX is relatively expensive so the ability to reuse material to create new samples would be of great use. The machining fines were able to be compacted in the die to produce recycled samples. The recycled samples had significantly smaller particle sizes than pristine [**Figure 44.1**]

44.3 Recent Progress

44.3.1 Material Characterization of Pristine MHE, Recycled MHE, and Other Materials of Interest

As part of a Round Robin experimental effort, LANL provided twelve pristine IDOX samples, where pristine means the sample has been produced from exact specifications covering ratios of the different IDOX particle sizes and the type of binder used, which is Estane 5703 and a proprietary nitroplasticizer. The aim of the round robin was to get measurements at typically difficult to achieve intermediate strain rates. The rates tested were 100/s, 10/s, and quasi-static 10^{-3}/s on the Gleeble thermomechanical simulator. The first samples tested, one at each strain rate, showed promising results [**Figure 44.2**] as the quasi-static yield strength matched that of a previously published set of data by Yeager et al [**Figure 44.3**]. They also followed an expected increase in strength with increasing strain rate [44.4].

Before continuing mechanical testing, the remaining pristine samples underwent x-ray computed tomography to collect particle distribution data for the modeling effort. Three samples were tested at each strain rate. The initial results of this secondary round of testing showed different behavior than observed in the initial samples [**Figure 44.2**], where the post-CT yield strengths were significantly higher [**Figure 44.4**]. After investigating x-ray effects on polymers, it was confirmed that there is a documented correlation between x-ray radiation and an increase in the strength [44.5]. Thus, a hypothesis was formed to explain the discrepancy in measurements; the x-ray radiation from computed tomography caused a change in the polymeric binder of the IDOX samples, resulting in an increased yield strength across strain rates.

To test this hypothesis, $CaCO_3$ samples in Kel-F binder (discussed in Section 44.3.2) were scanned for eight hours and compressed to failure in the Deben in-situ load frame. These results are contrasted against samples that did not undergo CT scanning [**Figure 44.5**]. Though the increase in yield stress is not as great as for the IDOX samples, it is still significant. Further testing will be performed to quantify differences in scan time and binder material used.

Finally, recycled IDOX samples were tested at the same strain rates as the round robin experiments. These resulted in strengths that were also much higher than the pristine samples [**Figure 44.6**] however this was in-line with expected results. The recycled samples have significantly smaller particle sizes on average than the pristine samples as seen in [**Figure 44.1**]. Past studies have shown an inverse relationship between particle size and yield strength [44.6]. These particle size interactions can be explained through an effective Hamaker constant, which typically defines van der Waals body-body interactions [44.7].

44.3.2 Formulation of New Surrogate Mock HE Samples

An investigation of alternative materials to IDOX was undertaken as part of an internship at LANL. Though IDOX is an ideal mock for PBX 9501 as it matches several different characteristics of HMX it is expensive and can be complicated to work with. With surrogate mocks, testing can be done quickly and inexpensively for situations where an exact match is not needed. A set of criteria was developed for determination of materials. Firstly, the density should be similar to that of IDOX/HMX within ± 0.5 g/m^3. Next, the material should be easily obtainable and ideally come ready for formulation. Currently, as received IDOX must be recrystallized and undergo particle sizing for the appropriate distribution of crystal sizes. Finally, the materials should be relatively inexpensive.

From these criteria, three materials were chosen for further investigation: calcium carbonate, alumina, and graphite flakes. After the materials were received, they were formulated according to mock HE specifications. The formulation process is as follows: a lacquer is formed by dissolving binder of choice in an appropriate solvent. The lacquer is mixed in with material at a ratio of 5% binder, 95% particulate to form a slurry. As the solvent evaporates,

the slurry is stirred continuously to prevent clumping. Finally, the mixture is allowed to fully dry for several hours. This powder can then be compacted into desired sample sizes. For each material type, four different binders were used for formulation: Estane 5703 plus nitroplasticizer (a lab proprietary additive for improved binder adhesion), Estane 5703, Estane 5716, and Kel-F. By using different binder types, their effects can be studied concurrently with the different particulate types and will be essential in further investigating the CT hardening effect.

44.3.3 In-situ Computed Tomography Experiments

The Zeiss Versa Xradia MicroCT cabinet can be used in conjunction with a Deben 5kN load frame. This allows for images to be taken pre- and post-compression, or, using several scans, quasi static deformation can be observed. This in-situ load frame is particularly useful for observing granular flow and particle movement within the samples. Additionally, LANL provided a ¼" die set that can be used within the Deben. In-situ pressing of the samples is of great importance to the modeling effort as it provides information on particle movement and interactions. For both pressing and compression testing, the Deben can perform "interrupted" or incremental movements with CT scans taken in between for a more complete view of particle movement before failure.

44.4 Plans for Next Reporting Period

- Interrupted in-situ pressing of IDOX and formulated F50
- Investigate other binder/material systems, including CT hardening effect on different binders
- Completion of Master's Thesis

44.5 References

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44.6 Figures and Tables

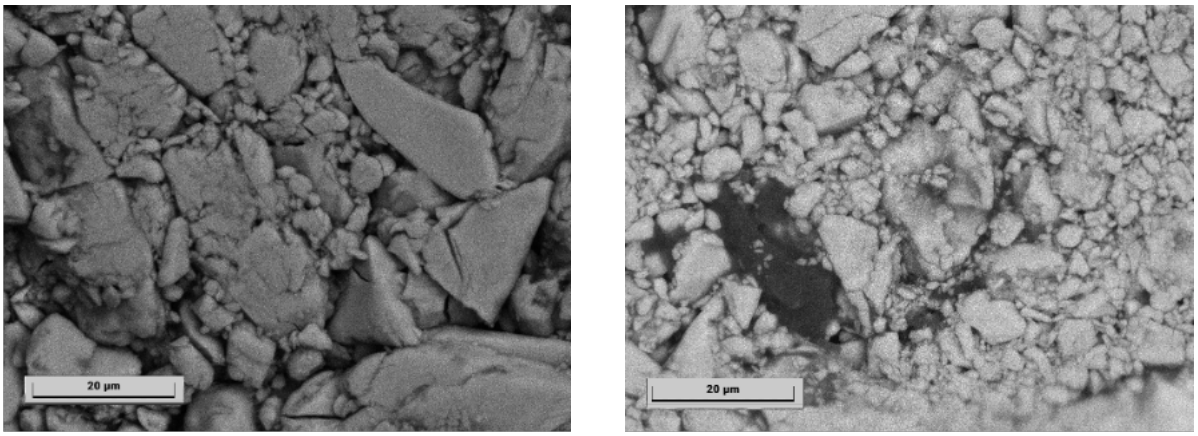

Figure 44.1: SEM imaging of pristine MHE (left) and recycled MHE (right) using TESCAN in secondary electron mode

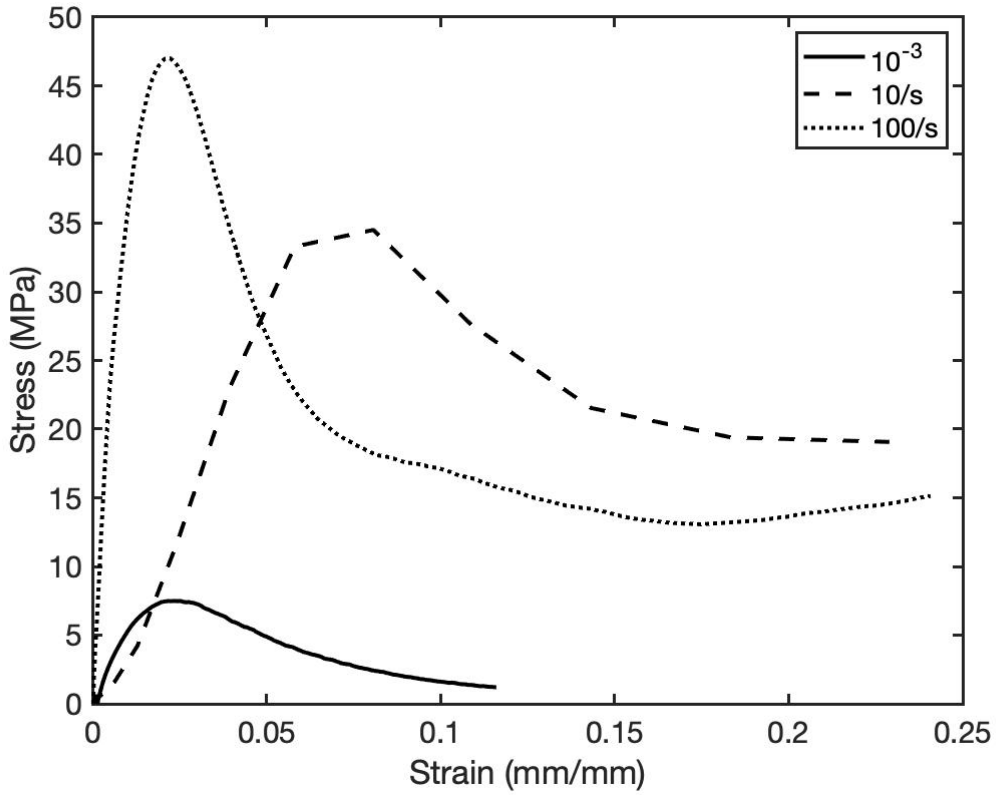

Figure 44.2: Stress vs. strain plots from compression testing of geometrically identical IDOX MHE samples (cylindrical, 0.25 inch high, 0.5 inch diameter) at the three indicated strain rates using a Gleeble thermomechanical simulator.

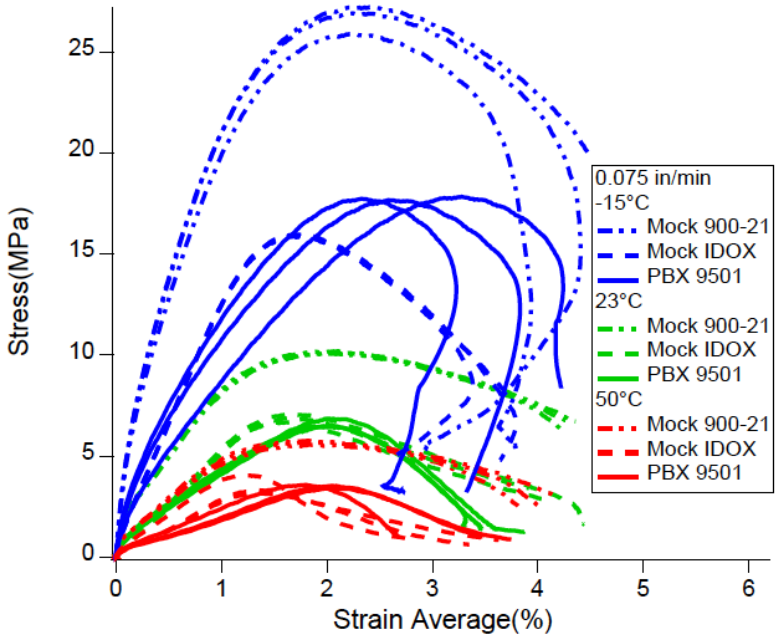

Figure 44.3: Quasi static stress vs strain for IDOX based mock, HMX high explosive, and mock 900-21. Yeager, John David, et al. *Development of a new density and mechanical mock for HMX*. No. LA-UR-18-25764. Los Alamos National Lab.(LANL), Los Alamos, NM (United States), 2020.

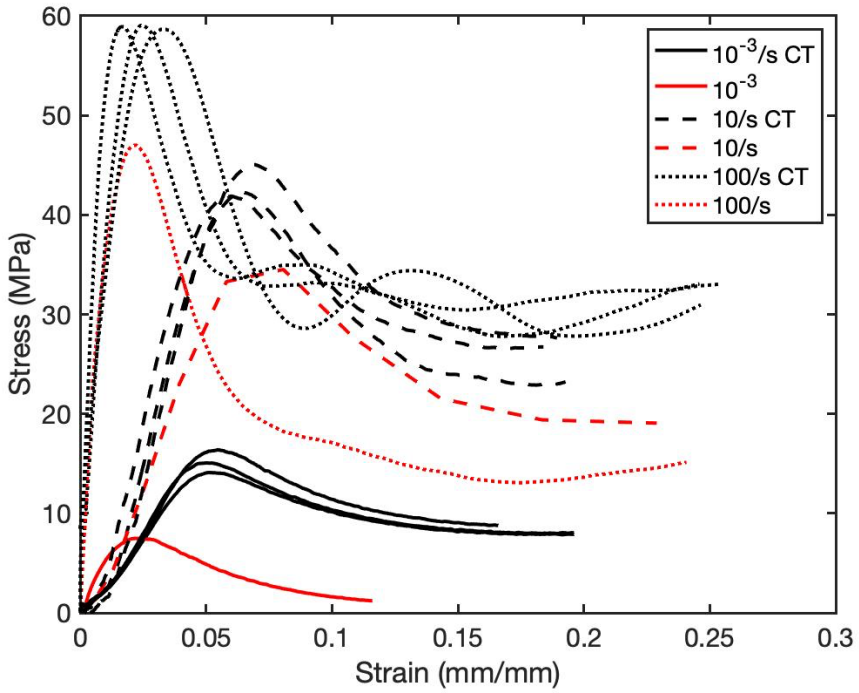

Figure 44.4: Stress vs. strain plots from compression testing of geometrically identical IDOX MHE samples (cylindrical, 0.25 inch high, 0.5 inch diameter) at the indicated strain rates showing difference in strength post CT imaging.

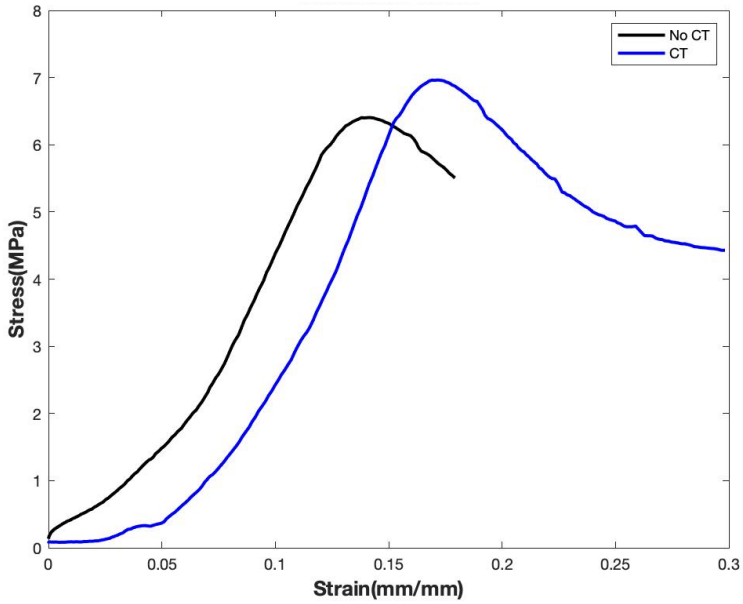

Figure 44.5: Stress vs. strain plots from compression testing of geometrically identical $CaCO_3$ samples (cylindrical, 0.125 inch high, 0.25 inch diameter) showing the difference in strength post CT imaging.

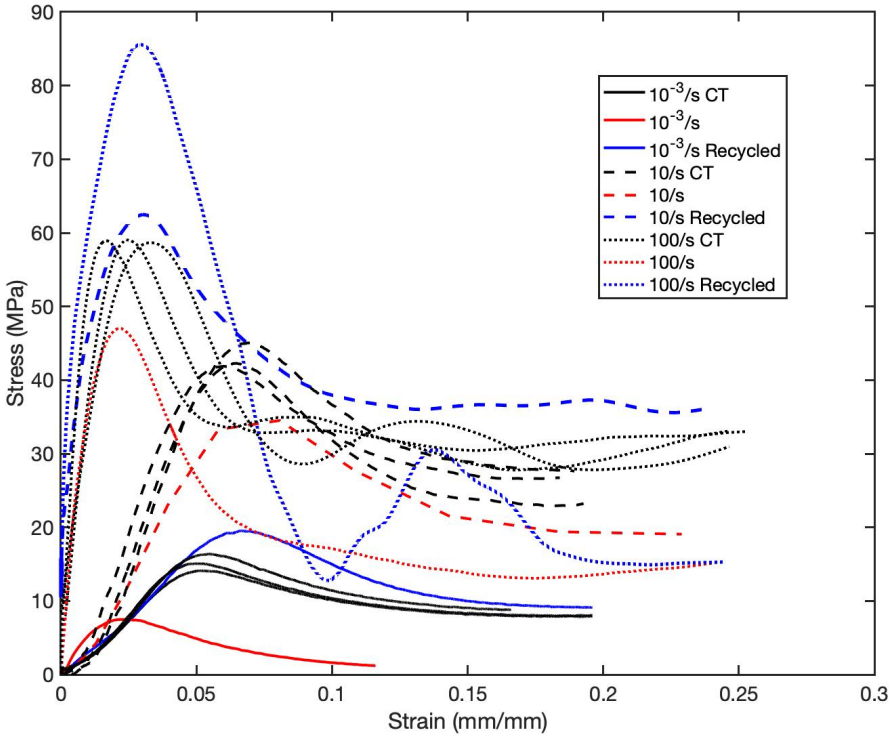

Figure 44.6: Stress vs. strain plots from compression testing of geometrically identical IDOX MHE samples (cylindrical, 0.25” high, 0.5” diameter) at indicated strain rates showing difference in strength post CT imaging and of recycled.