Equation-of-State Measured via X-ray Phase Contrast Imaging for Epon 828/DEA Epoxy

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Abstract. Epoxies are a broad class of polymer materials often used as adhesive, structural or binding materials. Epon 828 is an epoxy resin that can be polymerized with a variety of curing agents with the choice of curing agent potentially having an effect on the resulting epoxy polymer's material properties. In this study, the dynamic behavior of Epon 828 epoxy resin cured with diethanolamine (DEA) is investigated through a series of tamped Richtmyer-Meshkov instability (RMI) experiments measured with x-ray phase-contrast imaging. The measured shock and particle velocities are combined with data in the literature to calibrate Mie-Grüneisen equations-of-state (EOS) for portions and combinations of the collective dataset. The calibrated Mie-Grüneisen EOS are validated against particle velocity profiles extracted from published literature using the Eulerian hydrocode CTH. The Mie-Grüneisen EOS fit to only the tamped RMI experimental data presented here most closely follows the particle velocity profile in the published literature.

INTRODUCTION

Epoxies are polymer materials that are extensively used for engineering applications, including adhesive, structural, and binding applications for aircraft and automotive components [1, 2]. To predict how epoxy materials will behave in shock compression applications, numerical material models for the specific epoxy need to be calibrated and validated. Typically, the mechanical response of solid materials to shock compression is calculated using an equation of state (EOS) to define hydrostatic behavior and a strength model to define deviatoric behavior. Historically, polymers were believed to have minimal dynamic strength and were often simulated using only EOS models. However, a recent study by Bober, Lind, and Kumar [3] showed that simulations of material flow experiments using tungsten powder containing polymers could not be accurately reproduced without a pseudoplastic strength model for the polymer that generated shear resistances similar to solid metals [3]. Their results necessitated the investigation and calibration of dynamic properties for additional polymer materials [3]. In this study, a commonly used epoxy, Epon 828 [ThermoFisher, Product No. NC9610653] cured with diethanolamine [Sigma Aldrich, Product No. 398179] (Epon 828/DEA), is investigated using the tamped Richtmyer-Meshkov instability (RMI) method.

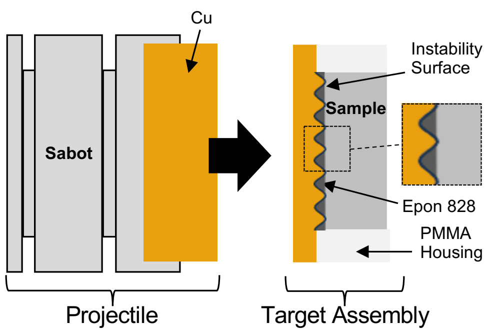
Epon 828 is a ubiquitous epoxy resin that can be polymerized with a variety of curing agents [4]. The EOS of various Epon 828-based epoxies have been previously investigated via planar impact experiments [5, 6]. Munson and May [6] studied the EOS of Epon 828 with three curing agents, the first being metaphenylenediamine, and two curing agent mixtures "D" and "Z" which were organic salt and aromatic mixtures. The Marsh [5] compendium of shock Hugoniot data contains an Epon 828 with an unspecified curing agent. Although this shock Hugoniot data exists in the literature, for many epoxies the defined EOS fails to distinguish the hardener used and in some cases the resin type. Properties of epoxies can vary with curing agent, even when using the same epoxy resin [7]. Therefore, it is important to expand upon the literature data by evaluating EOS's for specific resin-hardener combinations and to understand the dynamic response of these materials. A useful method for evaluating the dynamic response of materials is the tamped RMI method.

The tamped-RMI method generates an interfacial instability between shock compressed materials and calibrates material properties to the observed deformation behavior. Tamped-RMI experiments are performed by shock compressing a target assembly composed of two materials with a corrugated interface, typically in a sinusoidal pattern [8, 9]. Shock compression is typically applied via plate impact. The impacted and first shock compressed material is referred to as the driver, while the downrange material that the shock wave is transmitted into is referred to as the tamper. These experiments are typically performed with a negative Atwood number, meaning that the density of the driver material is greater than the density of the tamper material. This negative Atwood number allows the initial corrugation to invert under shock compression, jetting into the tamper material. This inversion and jetting behavior is dependent on a number of factors, including the applied stress, driver material strength, tamper material strength, Atwood number, and initial corrugation aspect ratio. By inserting the unknown material as the tamper and selecting a well-characterized impactor and driver material, the dynamic strength of the unknown material can be isolated and calibrated through iterative simulations [8].

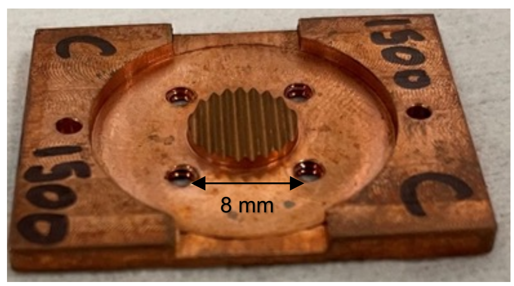
In order to determine the loading condition (pressure, density, temperature) associated with the strength values calibrated via tamped RMI, a calibrated and validated EOS must be defined for each material in the experiment. For dynamic simulations the Mie-Grüneisen EOS is commonly used. The Mie-Grüneisen equation is an incomplete EOS characterized by the Grüneisen coefficient that relates the pressure and volume of a solid at a given temperature [10]. The shock Hugoniot data along with the Grüneisen coefficient and a material density can be used to define a full EOS surface, i.e, the Mie-Grüneisen EOS.

In this study, three tamped RMI experiments were performed to investigate the dynamic response of Epon 828/DEA epoxy. The samples were impacted with a copper flyer plate launched from a powder gun at impact velocities ranging 1.2 to 2.2 km/s. The Epon 828/DEA dynamic response was tracked using x-ray phase-contrast imaging. A new calibrated Mie-Grüneisen EOS is calibrated, validated, and presented in this work for Epon 828/DEA.

EXPERIMENTAL DETAILS



(a)



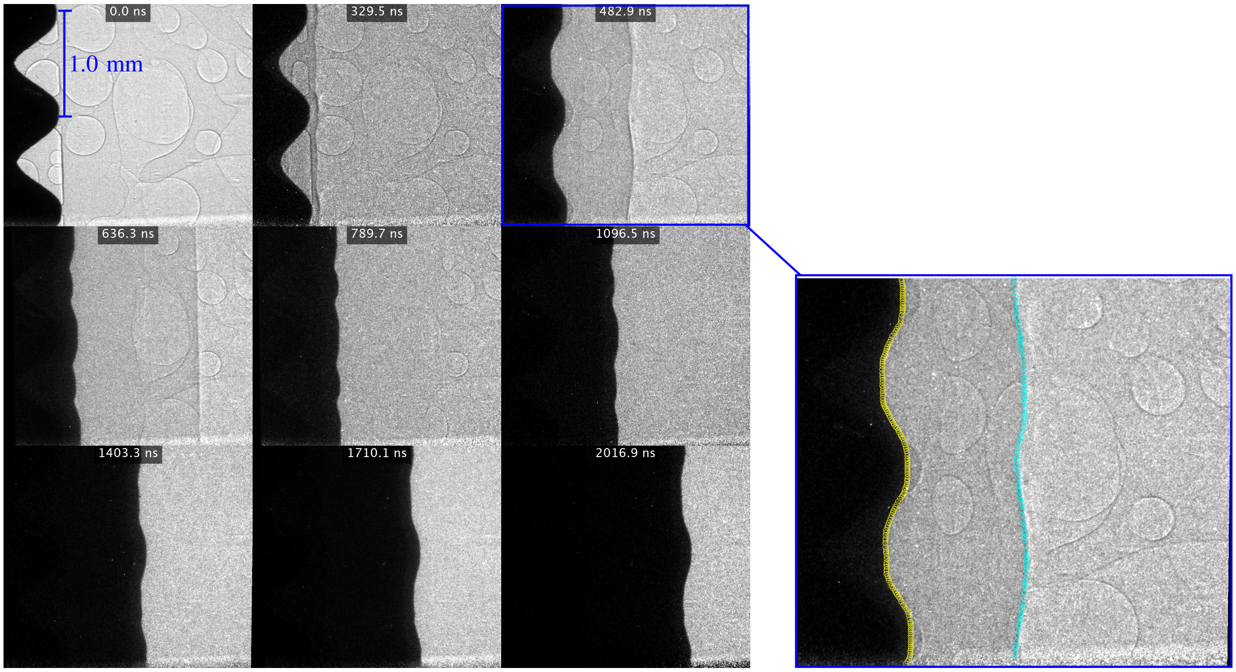
(b)

FIGURE 1. (a) Diagram of experimental setup. The dashed box indicates the camera field of view. X-ray path is out of the page. (b) Image of copper driver.

The experimental configuration for this study, as shown in Figure 1, consists of two main components: the copper driver and the sample, which in this case is Epon 828/DEA. The copper driver is 12.7 mm in width, 16.0 mm in length, and 2.0 mm in thickness with a sinusoidal corrugation 8 mm in diameter machined on the surface with various corrugation aspect ratios, $k\eta_0$. Directly against the corrugation surface is the Epon 828/DEA sample. To make the Epon 828/DEA sample, a large block of epoxy was cast by mixing a 100:12 ratio by mass of Epon 828 resin to DEA, degassing the mixture to remove air bubbles, and curing it in an oven at 70°C and standard pressure for 24 hours. The Epon 828/DEA sample was then cut from the block into a cylinder of 8 mm in diameter and 11 mm in height. These samples were then fixed to the copper driver by placing a PMMA housing with a thickness of 5.7 mm around the corrugated part of the driver and adding a layer of Epon 828/DEA to the corrugation to ensure there are no voids at the material interface. This additional layer of Epon 828/DEA was used to adhere the Epon 828/DEA cylindrical sample to the copper driver by using the same curing process as the block of Epon 828/DEA that the sample was cut from.

The samples were impacted with 3.9-4.1 mm thick, 10.0 mm diameter, oxygen-free high thermal conductivity (OFHC) copper flyer plates launched from a single-stage powder gun at the Dynamic Compression Sector (DCS) of the Advanced Photon Source facility (APS) at Argonne National Laboratory. Synchrotron radiation from the APS facility is used to perform in situ X-ray phase contrast imaging with a frame rate of 153.4 ns. The X-ray beam is focused directly over the corrugation of the sample to track the interface between the copper driver and the Epon 828/DEA sample in time. This beam lights the scintillator and a four-camera setup with 5x optic takes an image of the scintillator to produce a radiograph image. These radiograph images have a field of view of about 2.5 mm in width and 2.2 mm in height. The samples in this study were impacted at velocities of 1.617 km/s, 1.155 km/s, and 2.199

km/s, reaching stresses in the Epon 828/DEA samples of 6.28, 4.62, and 11.33 GPa respectively.



(a) **(b)**

FIGURE 2. (a) Experimental x-ray images obtained for shot 23-4-013. The copper driver appears as the dark region and the Epon 828/DEA appears as the light region with the direction of impact going from left to right. It is important to note that the bubbles that appear in these x-ray images are radial against the PMMA housing and neither present in the sample nor on the surface of the corrugation. (b) Image with extracted material interface (yellow circles) and shock wave (cyan diamonds) locations.

EQUATION OF STATE

To adequately capture experimental behavior in simulations, the EOS of the material must be defined. The Mie-Gruneisen EOS can be calibrated for a material by determining the shock velocity and the particle velocity during impact experiments and correlating a relationship between the two velocities. Although traditionally defined through the use of planar impact experiments, the shock velocity and particle velocity can still be evaluated with a corrugated surface as long as the driver-tamper interface and the shock wave can be tracked in time. There exist compendiums of Hugoniot EOS parameters for a number of materials [5, 11], however, for epoxies many defined EOS fail to distinguish which combination of epoxy resin and hardener were used. For this reason, it was important to establish an EOS for Epon 828/DEA that properly captures the material behavior under stresses in the 5-12 GPa range.

In this study, to define the EOS for Epon 828/DEA, experimental radiographs were used to identify the location of two key features: the interface between the copper driver and the Epon 828/DEA sample, and the shock wave location. This was done by hand-selecting the pixel coordinates of each feature in each frame using the `ImageJ` software. An example of the extracted contour locations of the interface between the two materials and the shock wave can be seen in Figure 3. To determine both the shock velocity, U_s, and the particle velocity, u_p, for the experiments, the extracted contour locations were used to find the average displacement in the radiograph frames and divided by the time between the frames. In addition to calculating U_s and u_p, the shock compressed density, ρ is calculated by multiplying the initial density of 1.186 g/cc by the ratio of the distance between the shock and the material interface in the pre-shock frame to the distance between the shock and the material interface in the shocked frame, i.e, volumetric compression.

Each experiment results in a value of shock velocity and particle velocity that can be plotted in U_s-u_p space and fitted with a linear trend line to obtain the representative EOS. The U_s-u_p values from this study and the EOS extracted from fitting the data can be found in Table I and Equation 1 respectively.

TABLE I. U_s-u_p data for Epon 828/DEA experiments performed.

Shot ID	$k\eta_0$	Impact Velocity (km/s)	Stress (GPa) [a]	U_s (km/s)	u_p (km/s)	ρ (g/cc)[a]	ρ (g/cc)[b]
23-4-007	1.00	1.617 ± 0.034	6.82 ± 1.45	4.79 ± 0.50	1.20 ± 0.07	1.58 ± 0.05	1.65 ± 0.04
23-4-013	1.50	1.155 ± 0.012	4.62 ± 0.72	4.40 ± 0.12	0.88 ± 0.11	1.48 ± 0.06	1.45 ± 0.10
23-4-015	0.75	2.199 ± 0.012	11.33 ± 1.40	5.61 ± 0.14	1.82 ± 0.16	1.76 ± 0.10	1.82 ± 0.08

[a] Calculated using Hugoniot equations and measured U_s, u_p, and ρ_0
[b] Evaluated using experimental contours to obtain the volumetric compression.

$$U_s = (1.496 \pm 0.116)u_p + (3.045 \pm 0.152) \tag{1}$$

Validation

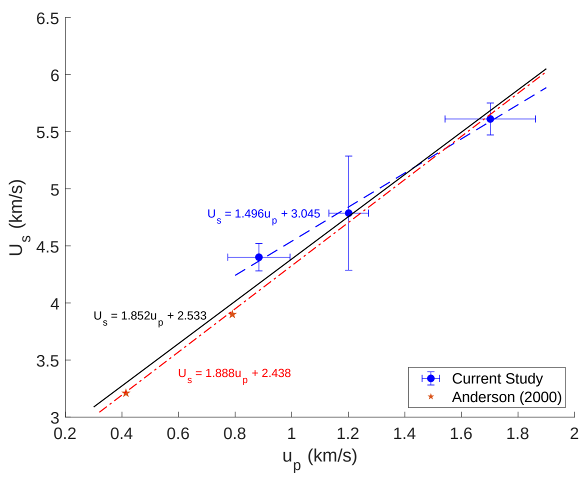


FIGURE 3. U_s-u_p data points for Epon 828/DEA from this study and from Anderson, Setchell, and Cox [12].

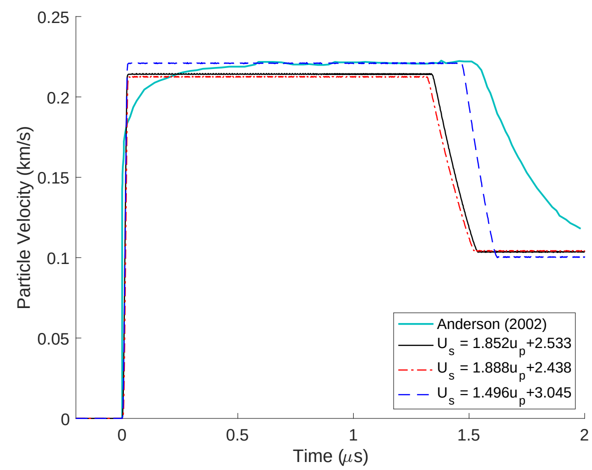
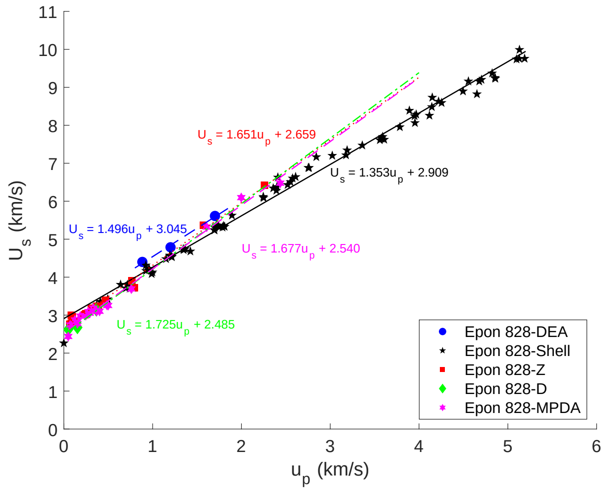


FIGURE 4. Comparison of various EOS to the experimental data of Anderson, Setchell, and Cox [13].

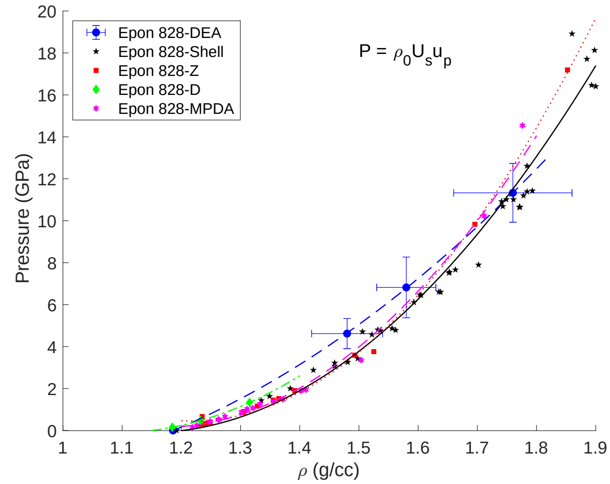
To ensure that the EOS model defined here was an accurate representation for Epon 828/DEA in the shock regime, the EOS model was validated against experimental data in the literature and compared to other possible representative EOS models. The plot shown in Figure 3, is a compilation of literature U_s-u_p points for Epon 828/DEA added to the data obtained from this study. Three separate linear trend lines were fit to the data, one for the points collected from this study (shown in blue in Figure 3), one that included the points from all the studies (shown in black), and the last removing what appeared to be an outlier point, which was the lowest stress shot in the present study (shown in red).

A one-dimensional simulation using the Eulerian hydrocode CTH that approximated the experimental geometry for Epon 828/DEA epoxy done by Anderson, Setchell, and Cox [13] was performed using each of the EOS models extracted from Figure 3 [12]. To accomplish this, a Mie-Grüneisen EOS was input by using the U_s-u_p EOS for the c_0 and s parameters and a Grüneisen gamma, Γ_0, of 1.13 [14]. The simulated profiles for each EOS were then compared to the experimental profile in velocity versus time space to evaluate which EOS best captures the experimental results of Anderson, Setchell, and Cox [13]. As seen in Figure 4, the EOS that most closely compared to the experimental results was the EOS derived from the current study, Equation 1, with a 0.48% difference between the simulation and the experimental steady-state particle velocity before the release wave.

To compare how this proposed EOS for Epon 828/DEA compares to other published data for Epon 828-based epoxy with various curing agents, it was plotted along with U_s-u_p data gathered from the Marsh [5] LASL compendium



(a)



(b)

FIGURE 5. (a) U_s-u_p relationships and (b) pressure vs shock compressed density for Epon 828 with varying curing agents using Epon 828-Shell from Marsh [5] and Epon 828-Z, Epon 828-D, and Epon 828-MPDA from Munson and May [6] and Epon 828-DEA from the current study.

and from Munson and May [6]. The data from the Marsh [5] LASL compendium is listed as Epon 828-Shell, however, it is unclear what "Shell" indicates, such as the brand for the Epon 828 resin or the type of curing agent. Munson and May [6] list the curing agents used in their study as metaphenylenediamine (MPDA), curing agent "Z" which is an active aromatic eutectic mixture of MPDA and phenylglycidyl ether, and hardener "D" which is an organic salt, the 2-ethyl hexoate salt of tridimethyl aminomethyl phenol. From Figure 5a, it can be seen that all the Epon 828 data have a similar trend regardless of the curing agent used as all the data points group closely together in U_s-u_p space. The EOS, however, do vary with the hardener used. The c_0 parameter for the Epon 828/DEA is close to that of Epon 828/Shell, but the slope has a significant difference. In addition, in Figure 5b, it can be seen that in pressure-density space all the data points fall along the same curve regardless of curing agent used. The EOS for Epon 828/MPDA and Epon 828/Z follow closely with each other while the EOS for Epon 828/D varies in slope. This means that while the shock Hugoniot data for Epon 828-based epoxies follow similar trends with different curing agents, using the EOS for the correct curing agent is preferable as the EOS parameters do vary with the curing agent used.

Uncertainty Characterization

Since the locations for the copper-Epon 828/DEA interface were hand selected from the x-ray images using `ImageJ` in this study, there is a level of uncertainty that comes with the recorded shock and particle velocities from which the EOS presented here is obtained. As seen in Figure 2b, there is a clear difference in intensity for the copper driver and the Epon 828/DEA in the x-ray images. For this reason, the image intensity was tracked along the length of the image to see when the intensity shift occurs from the copper driver to the Epon 828/DEA. The hand-selected locations were plotted along the image intensity and the error was determined to be the length of the intensity shift between the two materials compared to the hand-selected contour locations.

The minimum, maximum, and average length of the intensity shift across the set of frames was evaluated for each shot. This was then used for a Monte Carlo uncertainty evaluation for the resulting shock and particle velocities that utilize the hand-selected contour locations. The probability shape was defined as random because the true interface location between the two materials could be anywhere along the image intensity transition. The uncertainty was evaluated by generating a population of contour locations based off of the intensity shift and using the generated contour locations population to re-calculate the U_s-u_p data. The recalculated U_s and u_p for the complete population is used to determine the probability density of the associated velocities. From the resulting histograms, it is determined that the possible U_s-u_p values have a 95% confidence interval of $\pm$ 0.07 to 0.16 km/s.

CONCLUSIONS

In this study, we augment the available data for Epon 828/DEA by extracting the EOS in the 5 to 11 GPa pressure regime. This was accomplished with three tamped-RMI experiments that were performed using a single stage powder gun to shock compress Epon 828/DEA. The dynamic response of the experiments were captured using x-ray phase contrast imaging, which were then used to extract the driver-tamper interface and shock wave profiles. From these experimental profiles, the displacement across time was evaluated to obtain a shock and particle velocity for each experiment. The new U_s-u_p points were plotted together with other data points gathered from the literature and three linear relationships were explored by evaluating combinations of the data gathered in this study with literature data for Epon828-based epoxies. These three possible EOS models were tested by simulating a validation experiment performed with Epon 828/DEA by Anderson, Setchell, and Cox [13] and plotting the simulated particle velocity profiles against the experimentally measured data. The EOS fit to only the U_s-u_p points gathered in this study most closely followed the particle velocity profile published in [13].

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