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(U) A Grüneisen Equation of State for TPX: Application in FLAG

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Abstract

A Grüneisen equation of state (EOS) is developed for the polymer TPX (poly 4-methyl-1-pentene) within the LANL hydrocode FLAG. Experimental shock Hugoniot data for TPX is fit to a form of the Grüneisen EOS, and the necessary parameters for implementing the TPX EOS in FLAG are presented. The TPX EOS is further validated through one-dimensional simulations of recent double-shock experiments, and a comparison is made between the new Grüneisen EOS for TPX and the EOS representation for TPX used in the LANL Common Model.

1 Introduction

The development and testing of new materials under high-strain-rate conditions is often supplemented by computational modeling using hydrocode calculations. Specifically, if one desires to computationally model a high-strain-rate experiment one requires a computational representation of the properties of the material under investigation. Depending on the level of sophistication required for the calculation, material properties such as strength, fracture, compaction, etc. may or may not be included. However, at the most basic level each material must have an equation of state (EOS) to describes its equilibrium thermodynamic response. The EOS can take many forms, and one of the more commonly used forms is the Mie-Grüneisen EOS, a relatively simple EOS that does not specifically invoke an entropy term. Application of the Mie-Grüneisen EOS to the polymer TPX (poly 4-methyl-1-pentene) is explored in this work.

TPX is a polymeric material with a relatively low theoretical density and reasonable structural integrity. For these reasons TPX may be used in shock Hugoniot experiments for driving low pressure shocks into materials or for providing a low impedance material for achieving deep release states in a previously shocked material. As such, the ability to accurately capture its high-pressure response in hydrocode calculations is necessary. With a SESAME EOS for TPX not currently available in the LANL EOS libraries, the authors develop a Mie-Grüneisen EOS for TPX implementable in the hydrocode FLAG.[1] Development of the Mie-Grüneisen EOS is presented first, followed by application of the EOS to TPX. Finally, results from complex loading shock Hugoniot experiments are compared to hydrocode results using the newly developed Grüneisen TPX EOS.

2 Mie-Grüneisen EOS

If the Hugoniot of a material is known, the Mie-Grüneisen EOS can be used to calculate the material state in a condition off the principal Hugoniot. This is accomplished using the Mie-Grüneisen EOS in the form:

$$P - P_H = \gamma\rho(E - E_H) \tag{1}$$

where P and E are the pressure and energy in the material at the state of interest and the subscript H refers to the Hugoniot state. The remaining parameters, γ and ρ are the Grüneisen gamma and the shock compressed density, respectively. The Grüneisen gamma can take many forms, and for the purposes of the present application gamma is defined by:

$$\gamma = \frac{\gamma_0 + b\mu}{1 + \mu} \tag{2}$$

where γ_0 is the value of the Grüneisen gamma at ambient conditions, b is a constant to be determined, and μ is a measure of the compression, $\mu = \rho/\rho_0$ -1. Rearranging Eq. (1) with insertion of the definition of μ and recognizing that $E_H = \frac{1}{2}P_H(\frac{1}{\rho_0} - \frac{1}{\rho})$ gives:

$$P = P_H\left(1 - \frac{\gamma\mu}{2}\right) + \gamma\rho E \quad . \tag{3}$$

Equation (3) can be further expanded by using the conservation of momentum relation, $P_H = \rho_0 U_S u_p$, under the assumption that the initial pressure P_0 is negligible. The conservation of momentum equation introduces two new parameters, the shock velocity U_S and the material (or particle) velocity u_P. The shock

velocity represented in the cubic form is given by:

$$U_S = C_0 + S_1 u_P + S_2 \left(\frac{u_P}{U_S}\right) u_P + S_3 \left(\frac{u_P}{U_S}\right)^2 u_P \quad , \tag{4}$$

where C_0 is the bulk sound speed of the material at ambient conditions, and the parameters S_1, S_2, and S_3 are fitting parameters determined from the measured Hugoniot. To determine $S_{1,2,3}$ one must use the conservation of mass equation, $\rho_0 U_S = \rho(U_S - u_P)$, re-arranged in the following manner:

$$u_P = U_S \left(1 - \frac{1}{1+\mu}\right) \quad or \quad \frac{u_P}{U_S} = \left(1 - \frac{1}{1+\mu}\right) \quad , \tag{5}$$

which when substituted in to Eq. (4) results in a function for U_S in terms of compression:

$$U_S = \frac{C_0}{\left[1 - S_1\left(1 - \frac{1}{1+\mu}\right) - S_2\left(1 - \frac{1}{1+\mu}\right)^2 - S_3\left(1 - \frac{1}{1+\mu}\right)^3\right]} \quad . \tag{6}$$

When Eq. (5) is substituted back in to Eq. (3) through the conservation of momentum relation, this gives:

$$P = \underbrace{\rho_0 U_S^2}_{\text{(a)}} \underbrace{\left(1 - \frac{1}{1+\mu}\right)\left(1 - \frac{\gamma\mu}{2}\right)}_{\text{(b)}} + \underbrace{\gamma\rho E}_{\text{(c)}} \quad , \tag{7}$$

which must be further manipulated to achieve the desired form of the Mie-Grüneisen EOS. Beginning with segment (a) of Eq. (5) one arrives at:

$$\rho_0 U_S^2 = \frac{\rho_0 C_0^2}{\left[1 - S_1\left(1 - \frac{1}{1+\mu}\right) - S_2\left(1 - \frac{1}{1+\mu}\right)^2 - S_3\left(1 - \frac{1}{1+\mu}\right)^3\right]^2} \quad . \tag{8}$$

Distributing segment (b) results in:

$$\left[1 - \frac{\gamma\mu}{2} - \frac{1}{1+\mu} + \frac{\gamma\mu}{2(1+\mu)}\right] \quad . \tag{9}$$

Substitution of Eq. (2) and the definition of μ into segment (c) gives:

$$\left[\gamma_0 + b\mu\right]\rho_0 E \quad or \quad \left[\gamma_0 + b\mu\right]\epsilon \tag{10}$$

noting that $\epsilon = E\rho_0$ is the energy per initial unit volume ($V_0 = 1/\rho_0$). Substituting Eqs. (8), (9), and (10) into Eq. (7) yields:

$$P = \underbrace{\frac{\rho_0 C_0^2 \left[1 - \frac{\gamma\mu}{2} - \frac{1}{1+\mu} + \frac{\gamma\mu}{2(1+\mu)}\right]}{\left[1 - S_1\left(1 - \frac{1}{1+\mu}\right) - S_2\left(1 - \frac{1}{1+\mu}\right)^2 - S_3\left(1 - \frac{1}{1+\mu}\right)^3\right]^2}}_{\text{(d)}} + \left[\gamma_0 + b\mu\right]\epsilon \quad . \tag{11}$$

Multiplying both the numerator and denominator of segment (d) in Eq. (11) by $(1+\mu)^2/(1+\mu)^2$ results in the Mie-Grüneisen EOS in the desired form:

$$P = \frac{\rho_0 C_0^2 \mu \left[1 + \left(1 - \frac{\gamma_0}{2}\right)\mu - \frac{b}{2}\mu^2\right]}{\left[1 - (S_1 - 1)\mu - S_2 \frac{\mu^2}{1+\mu} - S_3 \frac{\mu^3}{1+\mu}\right]^2} + \left[\gamma_0 + b\mu\right]\epsilon \quad . \tag{12}$$

Equation (12) is the form of the Mie-Grüneisen EOS that is implemented in FLAG under the ".../gruneisen/" node, and describes the response of a material under compression. In tension the EOS takes the much simpler form:

$$P = \rho_0 C_0^2 \mu + \gamma_0 \epsilon \quad . \tag{13}$$

Through Eqs. (12) and (13) the equation of state of a material is fully defined within the Mie-Grüneisen framework. In the following section, this framework is applied to TPX to determine the appropriate EOS parameters necessary for implementing a Grüneisen EOS for TPX into FLAG.

3 TPX EOS Parameters

Implementation of a Grüneisen EOS for TPX into FLAG requires the definition of several thermodynamic quantities in addition to the material constants discussed above. The volumetric thermal expansion coefficient is taken from the manufacturer[2], α_V = 3.51 x10^{-4} /K, and the constant pressure specific heat from Ref. [3], C_P = 2,000 J/kg-K. The ambient sound speed and bulk density are taken from Marsh,[4] C_0 = 1.80 km/s and ρ_0 = 0.830 g/cm^3. The bulk modulus is then calculated from $K = \rho_0 C_0^2$ = 2.69 x10^9 kg/m-s^2. Using the identity that the bulk modulus is the inverse of the isothermal compressibility, $\beta = 1/K$ = -$(1/V)(\delta V/\delta P)_T$, one can also determine the constant volume specific heat, $C_V = C_P$ - $\alpha_V^2 VTK$ = 1,881 J/kg-K. The ambient Grüneisen parameter is then calculated from the thermodynamic identity:

$$\gamma_0 = \frac{\alpha_V V K}{C_V} = 0.60 \ . \tag{14}$$

Using the experimental data of Marsh,[4] the constants S_1, S_2, and S_3 can be determined using Eq. (6). The U_S-μ data is shown in Fig. 1, where a best fit to the data is given by the solid line with parameters S_1 = 2.246, S_2 = -1.30, and S_3 = 0.0. The density dependence of the Grüneisen parameter is assigned using the general form $\gamma_0\rho_0 = \gamma\rho$, which in terms of Eq. (2) is equivalent to setting $b = 0$. A summary of the Grüneisen EOS parameters for TPX for input into FLAG are given in Table 1

Table 1: TPX Grüneisen EOS FLAG input parameters.

$a = b = g_1$	c (cm/μs)	C_V ($cm^2/\mu s^2$-K)	g_0	r_0 (g/cm^3)	S_1	S_2	S_3	$tref$ (K)	$tzero$ (K)
0	0.180	1.881 x10^{-5}	0.60	0.830	2.246	-1.30	0	298	298

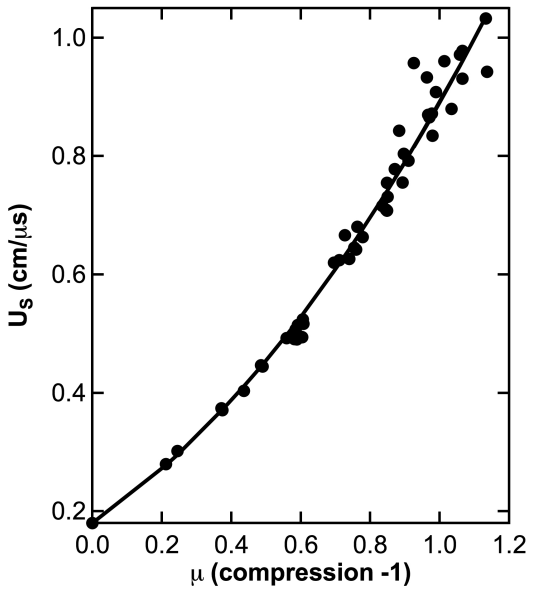

Figure 1: Data and corresponding parameter fit to U_S-μ response of TPX.

4 Re-Shock Experiment EOS Validation

The Grüneisen EOS for TPX is validated using results from a series of two re-shock experiments. In these experiments a projectile composed of a Lexan sabot, Ta backer, and TPX impactor was accelerated toward a stationary LiF window coated with ∼1 μm of Al using the LANL 50.8 mm two-stage gas gun. The material velocity at the TPX/LiF window interface was recorded using Photonic Doppler Velocimetry (PDV),[8] which when combined with the impact velocity and known Hugoniot of LiF can be used to calculate the Hugoniot state in TPX. Full details of the experiments can be found in Ref. [3]. The experiments were modeled in one-dimension using the LANL hydrocode FLAG. A schematic illustrating the one-dimensional problem set-up is given in Fig. 2. Initial conditions for the two experiments and corresponding one-dimensional calculations are given in Table 2.

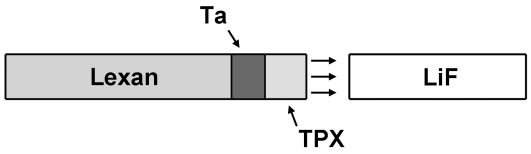

Figure 2: Idealized one-dimensional geometry of re-shock experiments simulated in FLAG.

To perform one-dimensional calculations on the geometry given in Fig. 2, material properties must be defined for each material. The Lexan sabot was defined using the polycarbonate (Lexan) SESAME EOS 7741, combined with the 'isocgy' elastic-perfectly plastic strength model. The shear modulus and yield

Table 2: Initial conditions for experiments and calculations.

	Shot (#)	Ta (cm)	TPX (cm)	LiF (cm)	u_D (cm/μs)
exp.	2s-447	0.2432	0.2998	1.2719	0.2146
calc.	2s-447	0.243	0.300	1.272	0.2146
exp.	2s-454	0.2451	0.3000	1.2726	0.2564
calc.	2s-454	0.245	0.300	1.273	0.2564

strength (flow stress) were taken from Rosenberg,[9] μ = 2.32 x10^9 kg/m-s^2 and σ_Y = 7.5 x10^7 kg/m-s^2, where the saturation and flow stress values were set to be equivalent. The melt temperature was further defined as 403 K. The LiF window was defined using the lithium fluoride SESAME EOS 7271, and was not assigned strength properties. The TPX EOS was defined using the Grüneisen EOS given in Table 1, and no strength model was applied.

For Ta, a Grüneisen EOS was generated in a manner similar to that for TPX. The volumetric thermal expansion coefficient was determined using an average of the linear thermal expansion coefficient for worked and annealed Ta from Hindert, α_V = 1.992 x10^{-5} /K.[5] The constant pressure specific heat at ambient conditions was C_P = 139.21 J/kg-K.[6] The ambient sound speed was taken from Marsh,[4] C_0 = 3.388 km/s. The bulk density was taken as the average from the Ta shock Hugoniot data reported in Marsh[4] and Mitchell and Nellis,[7] ρ_0 = 16.668 g/cm^3. Using the standard definition as above, the bulk modulus of Ta is calculated as K = 191.32 x10^9 kg/m-s^2. Using the previously defined constants, the constant volume specific heat is then C_V = 137.85 J/kg-K.

The experimental data of Marsh[4] and Mitchell and Nellis[7] is then used to fit the experimental constants for the Grüneisen EOS, Eq. (6), where the volume dependence of the Grüneisen parameter is assigned to the general form $\gamma\rho = \gamma_0\rho_0$. A summary of the Grüneisen EOS parameters for Ta are given in Table 3. The PTW strength model was defined for Ta,[10] where PTW parameters are those given in Ref. [11].

Table 3: Tantalum Grüneisen EOS FLAG input parameters.

$a = b = g_1$	c (cm/μs)	C_V ($cm^2/\mu s^2$-K)	g_0	r_0 (g/cm^3)	S_1	S_2	S_3	$tref$ (K)	$tzero$ (K)
0	0.3388	1.3785 x10^{-6}	1.659	16.668	1.113	0.392	0	298	298

The one-dimensional problem was defined with a mesh size of 5 μm, as was determined optimal from a previous study on flyer plate impact experiments.[12] A comparison of the actual and simulated target and sample thicknesses are given in Table 2. A tracer particle was imbedded in the LiF window 10 μm (2 cells) from the TPX/LiF interface, and recorded the Hugoniot state in the LiF window (pressure, P, material velocity, u_P, and density ρ) for comparison with the experimentally determined PDV wave profile. Upon initially running the calculations, a large numerical overshoot in material velocity was recorded in the tracer at the TPX/LiF interface due to wall heating of the cells near the TPX/LiF interface. To combat this issue, the artificial viscosity in the window was varied until an optimal condition was found. Values for the artificial viscosity parameters used in this study are given in Table 4.

Calculated results for the two experiments are shown with the measured wave profiles at the TPX/LiF

Table 4: Artificial viscosity parameters used in one-dimensional calculations.

	Lexan	Ta	TPX	LiF
q_1	0.2	0.2	0.35	0.40
q_2	1.0	1.0	2.0	6.5
q_{1n}	0.15	0.15	0.15	0.15

interface in Fig. 3, where fairly good agreement between the calculated and experimental results is observed. For the lower impact velocity shot, 2s-447, both the experimental arrival times and the steady state material velocities reached at each of the plateau regions align well with those from calculations. For the initial shock state, region "A", experimental and calculated material velocities overlap. For the first re-shock state "B" the calculated material velocity is approximately 1% higher than that measured experimentally, which reduces to ∼0.5% higher for the next re-shock state "C". By the time the final re-shock state "D" is reached both experiment and calculation lie on top of one another again. While the temporal arrival of the initial reduction in material velocity for the release state "E" is captured well for shot 2s-447, the subsequent release path differs between calculation and experiment, and may be a result of strength properties of the shocked Ta.[3]

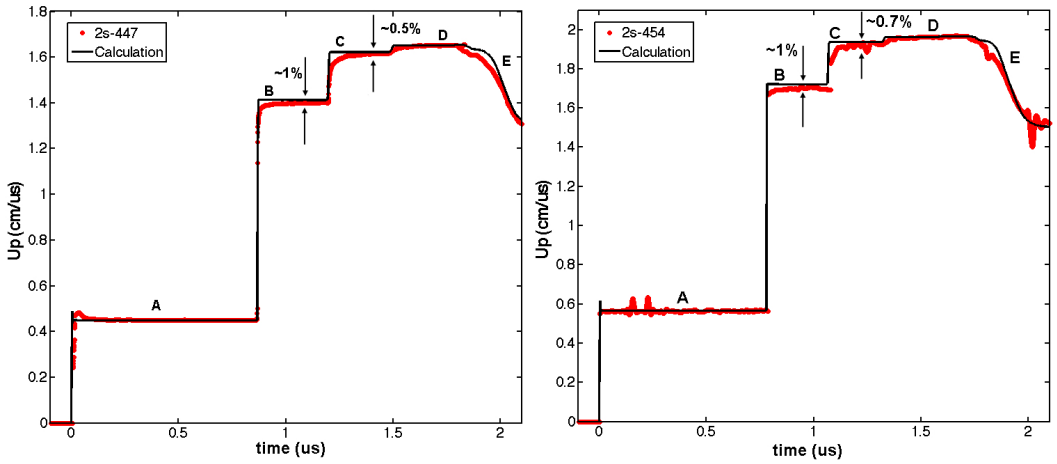

Figure 3: Experimental and calculated wave profiles at TPX/LiF interface for shots 2s-447 (left) and 2s-454 (right).

One obvious difference between the calculated and measured wave profiles for shot 2s-447 is the rounding near the leading edge of each of the measured peak velocity states. The shot report for 2s-447 states that the TPX impactor hit the LiF window at a tilt of 4.8 mrad, or 0.275°. Assuming the TPX impactor in the experiment has a diameter of 3.81 cm (1.5 in.), at an impact velocity of 0.2146 cm/μs this tilt results in a temporal difference at opposite ends of the impactor of 85 ns. This can consequently result in a dispersed wave front that may cause the observed rounding of the wave profiles at the leading edge of each of the peak states. The one-dimensional calculations are inherently free of tilt and exhibit sharp rises to the peak velocity states.

The higher velocity shot 2s-454 had slightly less tilt than shot 2s-447, and exhibits less rounding near the the leading edges of the plateau regions. Comparison of the measured and calculated wave profiles in Fig. 3 shows similarly good agreement in the plateau regions. The equilibrium state corresponding to the first shock "A" in the TPX is captured well with the prescribed Grüneisen EOS, with the subsequent calculated shocks "B" and "C" exhibiting similarly increased material velocities above the measured values. Measured jump-off times for each of the re-shock states correspond reasonably well with calculated values, as the calculated values for shocks "B" and "C" lead the measured values by approximately 8 and 15 ns, respectively. Similar to the lower impact velocity shot, shot 2s-454 also shows slight disagreement between experiment and calculation in the release portion of the wave profile.

Overall, relatively good agreement is observed between the initial and subsequent shocked states of TPX using the Grüneisen EOS for TPX defined in Table 1. Agreement is especially good for the initial shocked state, where subsequent re-shock states agree at the ∼1% or better level. Inspection of the current Common Model representation of TPX reveals that TPX is currently modeled using SESAME EOS 7230, the poly-tetra-deutero-ethylene (PTDE) EOS. The ambient density for PTDE is $\rho_0 = 1.093$ g/cm^3, significantly higher than the ambient density for TPX, $\rho_0 = 0.830$ g/cm^3. To account for the difference in initial density the current Common Model implementation for TPX employs a ramp treatment, where TPX at an initial density of 0.833 g/cm^3 is brought on to the PTDE EOS with a "Plastic Ramp" with ramp parameter $a = 0.004$. A comparison of the current Common Model PTDE EOS for TPX and the newly developed Grüneisen TPX EOS is given in Fig. 4. Inspection of Fig. 4 reveals that much better agreement is achieved with the measured wave profiles when the Grüneisen EOS is used for TPX rather than the PTDE EOS. As such, it is the recommendation of the authors that future TPX calculations employ the Grüneisen EOS for TPX given in Table 1, rather than the PTDE EOS with the ramp treatment, as is currently implemented in the LANL Common Model.

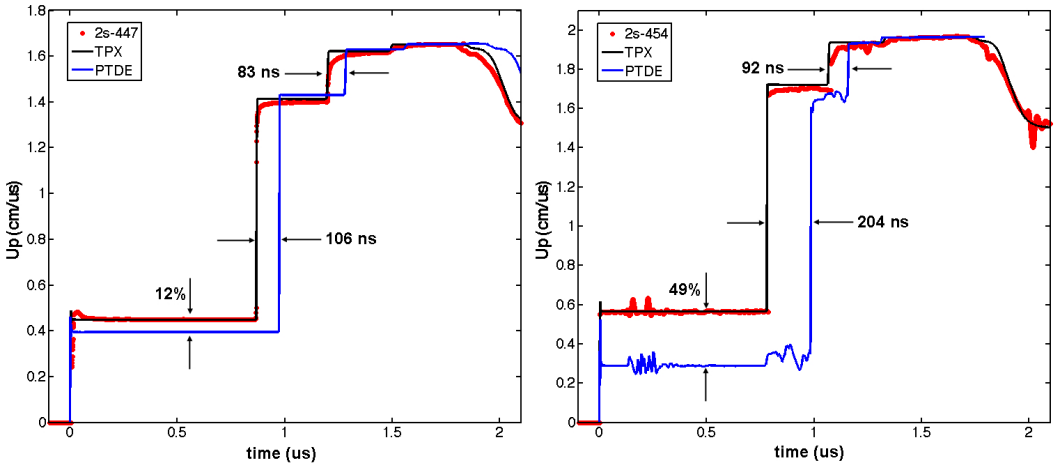

Figure 4: Experimental wave profiles at TPX/LiF interface compared to calculated profiles using TPX Grüneisen EOS and the PTDE EOS for shots 2s-447 (left) and 2s-454 (right).

5 Conclusions

A Grüneisen equation of state was derived for the polymeric material TPX (poly 4-methyl-1-pentene). A parameter set is developed for implementation of the Grüneisen EOS into the LANL hydrocode FLAG. The EOS is further validated against two high velocity impact experiments probing several re-shock states of TPX. One dimensional calculations of the experiments were able to capture the measured jump-off times of the re-shock states to within 15 ns for the highest impact velocity experiment, and plateau velocities of the initial and subsequent re-shock states with better than ∼1% accuracy for both experiments. Comparison of the current Common Model approach of modeling TPX with the PTDE EOS and a Plastic Ramp reveals that much better agreement with experimental data can be achieved if the newly developed Grüneisen EOS is used to model TPX.

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