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A Simple EOS for Linear Polytetradeuteroethylene

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ABSTRACT

A simple equation of state (EOS) for linear polytetradeuteroethylene (PTDE), with initial state density $\rho_0 = 1.093$ g/cm³, was generated and added to the T-4 Sesame EOS Library as material number 7230. This EOS reproduces the experimental shock Hugoniot data for PTDE and for isotopically scaled linear polyethylene.

INTRODUCTION

In this report, the generation of a simple equation of state (EOS) for linear polytetradeuteroethylene (PTDE) is presented. The EOS presented here reproduces the experimental shock Hugoniot data for PTDE and for isotopically scaled linear polyethylene, thus fulfilling the primary purpose of this EOS to describe the compression region.

METHOD

A simple EOS for PTDE was generated from various theoretical models using the $GRIZZLY^{1}$ computer code. (See the Appendix for a pedigree of GRIZZLY.)

For reasons of tractability, the models used to generate the simple EOS described here for PTDE do not <u>explicitly</u> treat PTDE as a polymer. However, experimental data for PTDE are used in these models, and thus the polymeric nature of PTDE is <u>implicitly</u> included in at least parts of the EOS.

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The repeat unit in the PTDE polymer chain is CD_2 . Therefore, PTDE was modeled as an average atom with an average atomic number of 8/3 and an average atomic weight <u>W</u> of 5.3469. An initial density $\rho_0 = 1.093 \text{ g/cm}^3$ at P = 0 ($P \sim 1$ bar) and T = 298.15 K was used.

The total EOS is a sum of cold curve (T = 0 K isotherm), thermal electronic, and nuclear (ion) contributions.

Cold Curve:

The cold curve in the compression regime from $\eta = 1$ to $\eta = 2.042$ (where $\eta = \rho/\rho_0$, where ρ is the density) was calculated from experimental shock Hugoniot data assuming a Mie-Grüneisen² EOS. Experimental shock Hugoniot data for PTDE^{3,4} and for ISM [linear polyethylene⁴ (Marlex) that has been isotopically scaled] were used. The η of 2.042 is the largest η for which there are experimental shock Hugoniot data.^{3,4} As seen in Fig. 1, the U_s vs U_p data for PTDE and ISM fall on the same curve.

The following fits to this shock Hugoniot data were used:

$$U_s = 2.441 + 1.990 U_p - 0.1560 U_p^2$$
, $U_p < 1.828$

and

$$U_s = 2.719 + 1.606 U_p - 0.02365 U_p^2$$
, $U_p > 2.342$,

where U_s is the shock velocity and U_p is the particle velocity; U_s and U_p are given in km/s. As with other polymers,⁵ two U_s vs U_p fits for PTDE were made, one below and one above the slight break in the U_s vs U_p curve at $U_p \sim 2$ km/s. (See Fig. 1.)

For $\eta \ge 2.042$, the cold curve was calculated using an analytic form⁶ for the Thomas-Fermi-Dirac (TFD) electronic model. At $\eta = 2.042$, the energy, the pressure, and the first derivative of the pressure were required to be continuous.

For $\eta \leq 1$, the cold curve was calculated with an analytic Lennard-Jones (LJ) form⁶ with the term FACLJ = 2 (corresponding to an r^{-6} attractive term, where <u>r</u> is the separation distance involved in the LJ pair potential). At $\eta = 1$, the energy, the pressure, and the first derivative of the pressure were required to be continuous.

The cohesive energy E_{coh} (the energy of vaporization of the solid at 0 K) used in this EOS for PTDE was calculated to be 0.302 MJ/kg, applying the estimation method of Bunn⁷ to the energy⁷ of vaporization of 680 cal/mole (at the normal boiling point) for the repeat unit, CH₂, of polyethylene.

At high temperatures, one expects the polymer to dissociate chemically. Thus, a dissociation energy (rather than a vaporization energy) might be more appropriate at higher temperatures. A dissociation E'_{coh} for a CH_2 repeat unit was calculated using bond strengths⁸ for a C-C single bond and for two C-H bonds. This E'_{coh} gave virtually the same numerical results for the total EOS in the compression region as did the E_{coh} of 0.302 MJ/kg, and gave similar qualitative results--but somewhat different numerical results--in the expansion region, especially for intermediate temperatures.

Thermal Electronic:

The thermal electronic contributions to the total EOS were calculated using a Thomas-Fermi-Dirac theory with an exchange constant of 2/3. This particular TFD theory is the same as that described in Ref. 9, except that the TFD theory used here for PTDE uses a local density approximation to the exchange.

Nuclear (Ion):

The nuclear (ion) contributions to the total EOS were calculated by a solid-gas interpolation scheme,⁶ which reduces to a Debye model at low temperatures and high densities and which reduces to an ideal gas at high temperatures or low densities.

The Grüneisen function γ and the Debye temperature θ at a given ρ were calculated from γ_0 and θ_0 using the Thompson formulas: 10

$$\gamma = \gamma_0 \xi + [2(1 - \xi)^2/3]$$

and

$$\theta = \theta_0 \eta^{2/3} \exp{\{\gamma_0(1 - \xi) - [(3 - 4\xi + \xi^2)/3]\}},$$

where $\xi = \rho_0 / \rho = 1/\eta$. An effective γ_0 of 0.4047 for PTDE was calculated using the Thompson formula and values of γ at various ρ calculated from a combination of off-Hugoniot data (sound speed and multishock measurements¹¹) and shock Hugoniot data^{3,4,11} for PTDE and ISM.

The θ_0 was calculated in the following manner: The lattice (intermolecular) contribution θ_{01} to θ_0 was calculated from

$$\theta_{01} = 444.12[(1 - 2\sigma)/(1 + \sigma)]^{1/2} c_0 [\rho_0/(W_{\lambda})]^{1/3}$$

with

$$\lambda = 2 + [(0.5 - \sigma)/(1 - \sigma)]^{3/2}$$

using $\rho_0 = 1.093 \text{ g/cm}^3$, the intercept c_0 (in km/s) of the given quadratic fit for U_s vs U_p shock Hugoniot data for PTDE, and a Poisson's ratio σ of 1/3; this equation⁶ can be derived from the usual relation between θ and the longitudinal and transverse sound speeds. For the quadratic fit for U_p < 1.828 km/s, $\theta_{01} = 248.4$ K; for the quadratic fit for U_p > 2.342 km/s, $\theta_{01} = 276.6$ K.

The intrachain (intramolecular) contribution θ_{02} to θ_0 was calculated in the following manner: For $\eta \le 1.493$ (corresponding to $U_D \le 1.828 \text{ km/s}$),

$$\theta_{02} = [(\theta_1 \theta_2 \theta_3 \theta_4 \theta_5 \theta_6 \theta_7 \theta_8 \theta_9)^{2/9} (\theta_{10} \theta_{11} \theta_{12})^{1/3}]^{1/3},$$

where these θ_i 's correspond to the following vibrational frequencies^{12,13} v_i 's (in cm⁻¹):

٧	: 2197	CD ₂ asymmetric
ν ₂	: 2192	stretch
vз	: 2102	CD ₂ symmetric
ν ₄	: 2088	stretch
^۷ 5	: 1146	CD ₂ bend
ν ₆	: 1093	\$
۷7	: 991	CD ₂ rosk
ν ₈	: 522	\$
ν9	: 916	CD ₂ twist

 v_{10} : 1249 C-C stretch

 ν_{11} : 1146 C-C stretch and C-C-C bend

 v_{12} : 522 C-C deformation.

The θ_{02} was calculated as a geometric mean¹⁴ of the θ_i 's, with each θ_i weighted according to the number of chemical bonds that involve the atoms in that vibrational mode v_i in the repeat unit of PTDE.

In a similar manner for $\eta > 1.572$ (corresponding to $U_p > 2.342 \text{ km/s}$),

$$\theta_{02} = [(\theta_{10}\theta_{11}\theta_{12})^{2/3}\theta_{14}]^{1/3} ,$$

where θ_{14} corresponds to the vibrational frequency $v_{14} = 3053 \text{ cm}^{-1}$ for D-D, which was calculated from the vibrational frequency¹⁵ of $v = 4315 \text{ cm}^{-1}$ for H-H using¹⁶ the ratio of the square roots of the masses of deuterium and hydrogen.

For $\eta \ge 1.572$, the polymer is being treated as a slightly different polymer, i.e., a crosslinked polymer with a repeat unit of C:

Here, deuterium atoms on neighboring polymer chains have come off as D_2 , and carbon-carbon single bonds have formed between neighboring chains, thus crosslinking the original polymer chains through side bonds. This very simplistic treatment is conservatively consistent with analyses¹⁷ of the products recovered from shock Hugoniot experiments on various polymers, which imply that the original polymer has undergone some chemical modification (which seems to involve some extent of irreversible dissociation) on the shock Hugoniot in the

regime above the break in the U_s vs U_p curve. That the crosslinked structure used here for $\eta > 1.572$ is only slightly different from the original PTDE structure used for $\eta < 1.493$ is highly consistent with the fact that, as with other saturated polymers,^{5,18} there is only a rather small change in the slope of the U_s vs U_p curve at the slight break at U_p ~ 2 km/s in the curve for PTDE. (See Fig. 1.) By the methods described above, θ_{02} was calculated to be 1680 K for $\eta < 1.493$ and 1956 K for $\eta > 1.572$.

The θ_0 was calculated as the geometric mean¹⁴ of θ_{01} and θ_{02} :

$$\theta_{o} = (\theta_{o1}\theta_{o2})^{1/2}.$$

Here, three normal modes have been assumed for the lattice vibrations and three normal modes have been assumed for the intrachain vibrations, thus leading to an equal weighting of θ_{01} and θ_{02} in θ_0 . In this manner, θ_0 was calculated as 646.0 K for $\eta \le 1.493$ and 735.6 K for $\eta \ge 1.572$.

Final EOS:

Tables of pressure, energy, and Helmholtz free energy for PTDE as a function of temperature and density were calculated in the manner described above, first using the input terms appropriate for $\eta < 1.493$ and second using the input terms appropriate for $\eta > 1.572$. GRIZZLY automatically sets the zero of energy at P = 0 and T = 298.15 K. The final tabular EOS is composed of the table values from the first EOS for $\eta < 1.493$ and of the table values from the second EOS for $\eta > 1.572$. This final EOS has been added to the T-4 Sesame EOS Library under material number 7230. The following kinds of pressure, energy, and Helmholtz free energy tables were added: 301 (total EOS), 304 (thermal electronic), 305 (ion, including zero point contributions), and 306 (cold curve, with no zero point contributions).

DISCUSSION

As seen in Fig. 1, the EOS described in this report for PTDE reproduces the experimental shock Hugoniot data^{3,4} for PTDE and ISM. Values of pressure, energy, and negative Helmholtz free energy for every other isotherm from the total EOS tables calculated for PTDE are plotted vs density in Figs. 2-4.

APPENDIX

PEDIGREE OF GRIZZLY

GRIZZLY¹ is a computer code put together by J. Abdallah (Group T-4) with contributions from J. D. Johnson (T-4) and B. I. Bennett (T-4). GRIZZLY uses subroutines taken, with appropriate modifications, from the computer codes PANDA,⁶ EOSCRAY, CANDIDE, and CHART D.¹⁰

PANDA is a computer code made by G. I. Kerley (then in T-4), using ideas and some coding from the computer code EOSLTS, developed by Bennett and modified by Bennett, Johnson, Kerley, and R. C. Albers (then in T-4). Some of the ideas and models in EOSLTS came from the following sources: the computer code SESAME, developed by J. F. Barnes and G. T. Rood (both then in T-4), and the CHART D radiation-hydrodynamic computer code¹⁰ of S. L. Thompson and H. S. Lauson (both of Sandia National Laboratories).

EOSCRAY is a version of EOSLTS. CANDIDE is a TFD computer code developed by D. A. Liberman (then in T-4) and modified by Bennett and Johnson.

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Fig. 1. Comparison of model and experimental shock velocity U_s vs particle velocity U_p on the Hugoniot. The solid curve is generated from the total EQS tables for PTDE with $\rho_0 = 1.093$ g/cm³. The experimental shock Hugoniot data^{3,4} are indicated by X's for PTDE and squares for isotopically scaled linear polyethylene.



Fig. 2. Some pressure vs density isotherms from the total EOS tables for PTDE. Pressure is in GPa; density is in $Mg/m^3 = g/cm^3$.



Fig. 3. Some energy vs density isotherms from the total EOS tables for PTDE. Energy is in MJ/kg; density is in Mg/m³.



Fig. 4. Some negative Helmholtz free energy vs density isotherms from the total EOS tables for PTDE. Helmholtz free energy is in MJ/kg; density is in Mg/m³.

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