Equations of state for solids at high pressures and temperatures from shock-wave data

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Abstract. This paper deals with the analytic derivation of a complete equation of state (EOS) for solids from shock-wave data in the range of pressures and temperatures, attained by detonation of chemical explosives. It is assumed that stresses behind the shock wave are isotropic, and the compressed materials behind the shock front are in thermodynamic equilibrium. A single Hugoniot curve determined from shock-wave experiments does not provide enough thermodynamic information to specify an EOS. The assumptions which, along with the shock-wave data, are used in the present work to determine a complete EOS are the Grüneisen assumption, the volume dependence of the Grüneisen parameter, and the Debye model for the specific heat. Consequently, the thermodynamic functions of the system under consideration are represented as a superposition of a cold (0 K) term corresponding to the cold compressed state and a second one corresponding to the thermal vibrations of atoms in the crystal lattice about their mean positions. A differential equation is derived for the cold isotherm of the internal energy, using the Mie-Grüneisen model and the shock Hugoniot as a reference curve. An analytic solution to the equation is obtained. Based on this solution, the complete, the caloric, the thermal and the incomplete EOS are determined. Also, an equation is derived for the shock temperature along the Hugoniot curve.

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Introduction

The study of shock-wave propagation in solids has added significantly to our understanding of physical processes which take place at high pressures, high temperatures and very short times. One particular aspect of these studies is to determine the equations of state (EOS) for different materials from shock-wave data. Shock-wave experiments, aimed at EOS investigations, render it possible to extend the range of pressure-volume data beyond the region that can be reached with conventional static pressure experiments and to use much bigger specimens of the investigated material.

Equations of state of matter, obtained both theoretically and experimentally are of immense current importance in the basic and the applied sciences. They describe the dependence of thermodynamic properties of matter on its microscopic internal structure and have a variety of applications. These include studies of the state of matter in the Earth’s and planetary interiors, shock-induced chemical reactions and polymorphic phase transformations, hypervelocity impact, powder compaction or other applications as well as their computer simulation based on hydrodynamic models. In principle, all the thermodynamic properties of a system can be predicted once precise knowledge of the complete EOS becomes known. But a single Hugoniot curve determined from shock-wave experiments does not provide enough thermodynamic information to specify an EOS. The assumptions which, along with shock-wave data, are used in this work to determine a complete EOS are the Grüneisen assumption (1), the volume dependence of the Grüneisen parameter (2), and the Debye model for the specific heat (3). Also, it is assumed that stresses behind the shock wave are isotropic, and the compressed materials behind the shock front are in thermodynamic equilibrium.

Therefore the objective of the work presented herein is to obtain a complete EOS for solids from shock-wave data using the Debye - Mie-Grüneisen model and the shock Hugoniot as a reference curve, and to establish the thermodynamic properties of a system with such an EOS. Also, an equation for the shock temperature along the Hugoniot curve will be derived.

1. General formulation

Formally, the equation of state is a functional relationship among the thermodynamic variables for a system in equilibrium. One of the most frequently used thermodynamic relations is the thermal EOS, or just the EOS. For a simple thermodynamic system it has the form

\[ f(P, V, T) = 0. \]  

(1.1)

It represents a surface in the \( PVT \) space. The \( PV \) isotherm (\( T = \text{const.} \)), isentrope (\( S = \text{const.} \)) and the shock Hugoniot (the locus of all possible states that can be reached by using a single shock from a given initial state) are particular curves on this surface.
To obtain the form of $f$, it is convenient to derive the Helmholtz free energy of the system

$$F = E - TS$$

and compute $P$ as the volume derivative

$$P = -(\partial F/\partial V)_T. \hspace{1cm} (1.3)$$

It is clear that $F$ depends upon the microscopic structure of the solid under consideration, which would vary as a function of volume and temperature. At different temperatures and densities, the corresponding region of matter will be dominated by different interactions.

In view of the above, we may write the free energy as a superposition of terms appropriate to various physical phenomena (Grüneisen’s assumption [1])

$$F(V, T) = F_c(V) + F_{vib}(V, T) + F_e(V, T) \hspace{1cm} (1.4)$$

Here $F_c$ is the configurational free energy at 0K. $F_{vib}$ corresponds to the contribution of the zero and thermal vibrations of the ions of the crystal lattice. $F_e$ is the conduction electron thermal excitations contribution.

Expressions for the internal energy, pressure and other thermodynamic functions are similar to Eq.(1.4):

$$P = P_c + P_{vib} + P_e, \hspace{0.5cm} E = E_c + E_{vib} + E_e. \hspace{1cm} (1.5)$$

The contribution of the conduction electrons thermal excitations manifests itself at temperatures $\geq 10^4 K$ and pressures $\geq 10^2 GPa$ [2]. At lower temperatures and pressures it may be neglected and Eqs.(1.4) and (1.5) take the form

$$F = F_c + F_{vib}, \hspace{0.5cm} P = P_c + P_{vib}, \hspace{0.5cm} E = E_c + E_{vib}. \hspace{1cm} (1.6)$$

The vibrational terms are sums of the quantum zero-degree and the thermal vibrations of the crystal lattice. Then Eqs.(1.6) may be written as

$$F(V, T) = F_c(V) + F_0(V) + F_{IT}(V, T), \hspace{0.5cm} E(V, T) = E_c(V) + E_0(V) + E_{IT}(V, T), \hspace{0.5cm} P(V, T) = P_c(V) + P_0(V) + P_{IT}(V, T). \hspace{1cm} (1.7)$$

$F_0$, $E_0$ and $P_0$ are the quantum zero-degree contributions. They are functions of volume only, like the configurational terms. $F_{IT}$, $E_{IT}$ and $P_{IT}$ are the lattice thermal contributions. They depend on volume and temperature.

The pressures and temperatures attained by detonation of chemical explosives in contact with the specimen or explosively accelerated flyer plate methods do not exceed $10^4 K$ and $10^2 GPa$. These techniques are the most accessible to the general scientific community and because of that the most frequently used ones. The behaviour of a solid within this range of pressures and temperatures is determined by the lattice thermal vibrations. That is why it is often called the phonon region. It looks narrow on the background of the highest temperatures and pressures, investigated by contemporary physics. But in addition to being easily accessible, it covers a wide variety
of processes and phenomena of interest to science and technology. Those mentioned in
the introduction are to name but few. Consequently, we shall aim our considerations at
the phonon region and seek the explicit form of Eqs.(1.7).

From Eq.(1.2) and the fundamental thermodynamic identity
\[ dE = -PdV + TdS \]
it follows
\[ E_c = F_c \quad \text{and} \quad P_c = -dF_c/dV = -dE_c/dV. \quad (1.8) \]
The specific form of Eqs.(1.8) will be derived from the Mie-Grüneisen equation using
the shock Hugoniot as a reference curve.

The calculation of Helmholtz free energy components \( F_0(V) \) and \( F_{IT}(V,T) \) will be
performed within the framework of the Debye model for the specific heats.

2. Calculation of the zero-degree Kelvin isotherm from shock-wave data

The shock-wave methods for deriving the cold isotherm are based on the measurement of
the Hugoniot curve. The quantities directly measured are the kinematic parameters of
the shock wave - the shock front velocity \( u_s \) and the particle velocity in the compressed
region \( u_p \). The relation \( u_s(u_p) \) is the shock Hugoniot. Most substances in the absence
of phase transitions have a linear shock Hugoniot [3]
\[ u_s = c_0 + su_p \quad (2.1) \]
in a wide range of particle velocities. The Hugoniot intercept, \( c_0 \), and the slope, \( s \), are
determined from the data by the method of least squares. If rigidity effects and possible
low pressure phase changes are neglected, the intercept should correspond to the velocity
of an infinitesimal pressure pulse, or the bulk sound speed, \( c_0 =[(\partial P/\partial \rho)_S]^{1/2} \) (at \( P = 0 \)). Since the slope is linearly related to the pressure derivative of the adiabatic bulk
modulus, \( (\partial B/\partial P)_S \), a linear \( u_s-u_p \) Hugoniot then reflects a nearly linear dependence of
\( B_S \) on the pressure.

The transition from kinematic \( (u_s,u_p) \) to thermodynamic \( (P,V,E) \) variables is done
using the laws of conservation of mass, momentum and energy across the shock front [3]
\[ \rho_0 u_s = \rho(u_s - u_p), \quad (2.2) \]
\[ P - P_0 = \rho_0 u_s u_p, \quad (2.3) \]
\[ \left[ (E - E_0) - \frac{u_p^2}{2} \right] \rho_0 u_s = P_0 u_p. \quad (2.4) \]
Here \( E, P \) and \( \rho \) are the specific internal energy, the pressure and the density behind
the shock front, and \( E_0, P_0, \rho_0 \) are the values of these quantities ahead of the shock front.
From Eqs.(2.2) and (2.3) we obtain
\[ u_s = V_0 \left[ (P - P_0)/(V_0 - V) \right]^{1/2}, \quad (2.5) \]
\[ u_p = \left[ (P - P_0)(V_0 - V) \right]^{1/2}. \quad (2.6) \]
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Solving Eqs. (2.5) and (2.6) for $P$ gives

$$P = P_0 + \frac{1}{V_0} u_s u_p, \quad V = V_0 \left( \frac{u_s - u_p}{u_s} \right). \quad (2.7)$$

Equations (2.7) give the relationship between the kinematic variables $u_s$, $u_p$ and the thermodynamic variables $P$ and $V$.

Substituting with the right-hand sides of Eqs. (2.5)-(2.6) in Eq. (2.4) gives the Hugoniot equation of energy

$$E - E_0 = (P + P_0)(V_0 - V)/2. \quad (2.8)$$

that defines all states on the $(E-P-V)$ surface that can be reached from an initial state $(E_0, P_0, V_0)$ by a single shock.

If the linear $u_s-u_p$ relation holds, the Rankin-Hugoniot equations (2.7) and (2.8) can be used to express pressure and energy as functions of volume along the Hugoniot by the following convenient analytic expressions (with $P_0$ and $E_0$ taken to be zero at ambient conditions)

$$P_H = \frac{\rho_0 c_0^2 \varepsilon}{(1 - \varepsilon s)^2}, \quad (2.9)$$

$$E_H = \frac{c_0^2 \varepsilon^2}{2 (1 - \varepsilon s)^2}, \quad (2.10)$$

$$\varepsilon = 1 - V/V_0 = 1 - \rho_0/\rho. \quad (2.11)$$

Equations (2.9)-(2.10) and the values of $c_0$ and $s$ summarize all the experimental thermodynamic information which is available from shock-wave measurements.

To compute the cold compression curve we shall employ the Mie-Grüneisen equation in the form

$$P(V,T) - P(V,0) = \frac{\gamma(V)}{V} [E(V,T) - E(V,0)], \quad (2.12)$$

where $\gamma(V)$ is the Grüneisen parameter. Equations (2.9,2.10) give the pressure $P_H$ and the specific internal energy $E_H$ on the shock Hugoniot. Since it connects equilibrium thermodynamic states we can write

$$P_H(V) - P(V,0) = \frac{\gamma(V)}{V} [E_H(V) - E(V,0)]. \quad (2.13)$$

We shall assume that $\gamma$ is a function only of volume and, specifically, that the product $\rho\gamma$ is constant. Experimental work on a number of materials, as well as theoretical considerations, indicate this to be an adequate approximation. With this assumption, $\gamma(V)$ is given by

$$\gamma(V)/V = \gamma_0/V_0, \quad (2.14)$$

where $\gamma_0$ is the thermodynamic value at standard conditions given by $\gamma_0 = 3\alpha c_0^2/C_p$, $\alpha$ is the thermal expansion coefficient, $c_0$ the sound speed, and $C_p$ the specific heat at constant pressure.
At $T = 0K$ $F = E$ and from Eqs. (1.8) it follows

$$P_c(V) = P(V, 0) = -dE_c(V)/dV.$$  

(2.15)

If we substitute with Eqs. (2.9), (2.10), (2.15) in the left-hand side of Eq. (2.13), we obtain a differential equation for the zero Kelvin isotherm of the specific internal energy $E_c(\varepsilon) = E(\varepsilon, 0)$:

$$dE_c(\varepsilon)/d\varepsilon - \gamma_0 E_c(\varepsilon) - \frac{c_0^2(1-\gamma_0/2)}{(1-s\varepsilon)^2} = 0,$$

(2.16)

$$E_c(\varepsilon = 0) = E_{00}.$$ 

This is a first order linear inhomogeneous ordinary differential equation. The functional form of $E_c(\varepsilon)$ may be obtained by integrating Eq. (2.16). Unfortunately, the solution cannot be given by combinations of elementary functions. It is possible, however, that it can be approximated by a power series in $\varepsilon$ within high precision. In this case the cold compression curve $E_c(\varepsilon)$ can be written as the following power series in $\varepsilon$

$$E_c(\varepsilon) = \sum_i E_{0i}\varepsilon^i,$$

(2.17)

where $\varepsilon$ is the dimensionless volume given by Eq. (2.11). $E_{00}$ is obtained from the assumed reference state and the remaining coefficients to the fifth order in $\varepsilon$ are given by the expressions:

$$E_{01} = \gamma_0 E_{00}, \quad E_{02} = \frac{1}{2} \left(c_0^2 + \gamma_0^2 E_{00}\right), \quad E_{03} = \frac{1}{6} \left(4sc_0^2 + \gamma_0^3 E_{00}\right),$$

$$E_{04} = -\frac{1}{24} \left[2 (\gamma_0 - 9s) c_0^2 s - \gamma_0^4 E_{00}\right],$$

$$E_{05} = -\frac{1}{120} \left[2 \left(\gamma_0^2 + 9\gamma_0 s - 48s^2\right) c_0^2 s - \gamma_0^5 E_{00}\right].$$

(2.18)

From Eqs. (2.13) and (2.17) we can derive an expression for the zero Kelvin isotherm of the pressure:

$$P_c(\varepsilon) = \sum_i P_{0i}\varepsilon^i.$$ 

(2.19)

The coefficients $P_{0i}$ and $E_{0i}$ are not independent. A relation exists between them which is given by

$$P_{0i} = (i+1)E_{0i+1}/V_0.$$

(2.20)

3. Contribution of the quantum zero Kelvin and lattice thermal vibrations to the EOS

In accordance with the quasiharmonic approximation the vibrational energy levels of a crystal lattice with $N$ atoms may be treated as the energy levels of a system of $3N$
independent linear harmonic oscillators. For the logarithm of the partition function of such a system we can write

$$\ln Z = \sum_{i=1}^{3N} \ln z_i = \sum_{i=1}^{3N} \ln \frac{\exp (-h\nu_i/2kT)}{1 - \exp (-h\nu_i/kT)},$$

(3.1)

where $z_i$ is the partition function of the $i$-th oscillator.

In the Debye model [7] the crystal lattice is replaced by an isotropic elastic medium with a continuous dispersion law function. Further it is assumed that the magnitude of frequency in such a system does not exceed a certain boundary value, the Debye frequency $\nu_D$, chosen so that the number of the independent lattice oscillations is equal to the total number $3N$ of the lattice degrees of freedom. It follows from this approximation that we can replace the summation in Eq.(3.1) by integration. Let us omit the index $i$ of the oscillations. Then

$$\ln \frac{\exp (-h\nu/2kT)}{1 - \exp (-h\nu/kT)}$$

is $\ln z$ for all oscillators in the frequency range between $\nu$ and $\nu+d\nu$. The number of these oscillators is equal to $g(\nu)d\nu$, where

$$g(\nu) = \frac{9N\nu^2}{\nu_D^3}$$

(3.2)

is the density of distribution of oscillators among frequencies [7]. We introduce the Debye temperature $\theta_D = \hbar\nu_D/k$, which depends on volume and is specific for each substance, and the new integration variable $x = h\nu/kT$. Then Eq.(3.1) takes the form

$$\ln Z = -\frac{9}{8}N (\theta_D/T) - 9N \frac{1}{(\theta_D/T)^3} \int_{0}^{\theta_D/T} x^2 \ln [1 - \exp (-x)] dx.$$

(3.3)

Integration by parts of Eq.(3.3) gives

$$\ln Z = -\frac{9}{8}N (\theta_D/T) - 3N \ln [1 - \exp (-\theta_D/T)] + 3N \frac{1}{(\theta_D/T)^3} \int_{0}^{\theta_D/T} \frac{x^3 dx}{e^x - 1}.$$

The last equation can be written in a more compact form if we introduce the function

$$D(z) = \frac{3}{z^3} \int_{0}^{z} \frac{x^3 dx}{e^x - 1},$$

which is known as the Debye function and $z = \theta_D/T$. Then the logarithm of the partition function takes the form

$$\ln Z = -\frac{9}{8}N (\theta_D/T) - 3N \ln [1 - \exp (-\theta_D/T)] + ND(\theta_D/T).$$

(3.4)

From Eq.(3.4) and the thermodynamic identities

$$F = -kT \ln Z, \quad E = F - T(\partial F/\partial T)_V,$$

$$P = -(\partial F/\partial V)_T, \quad C_V = (\partial E/\partial T)_V$$

we can derive expressions for the specific free and internal energy, the pressure and the specific heat:

$$F_{\text{vib}}(T, V) = \frac{9}{8}Nk\theta_D + NkT \{3 \ln [1 - \exp (-\theta_D/T)] - D(\theta_D/T)\},$$
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\[ E_{vib}(T, V) = \frac{9}{8} N k \theta_D + 3 N k T D(\theta_D/T), \]
\[ C_V = 3 N k \left[ D(\theta_D/T) - (\theta_D/T) D'(\theta_D/T) \right], \]

where \((9/8)Nk\theta_D\) is the contribution of the quantum zero oscillations in terms of the Debye model, and \(D'\) is the derivative of \(D\) with respect to \(\theta_D/T\).

In order to derive \(P_{vib}\) it is necessary to compute the volume derivative of \(\ln Z\). The partition function does not depend explicitly on volume but through the dependence of the energy levels on it. It is not possible to obtain this relation from first principles. This imposes the use of various approximations. In the case of a solid the volume dependence of the energy levels is reduced to volume dependence of the lattice frequencies. This relation cannot be obtained from first principles as well. According to Grüneisen

\[ d \ln \nu_j / d \ln V = -\gamma \quad (j = 1, 2, ..., 3N), \]

(3.5)

where \(\nu_j(V)\) are lattice frequencies, and \(\gamma\) is a positive quantity, one and the same for all the \(3N\) normal modes of the crystal lattice. It is assumed that these frequencies, and hence, \(\gamma\), do not depend on temperature, but on volume only. This is sometimes referred to as the quasiharmonic approximation.

Equation (3.5) is the statistical definition of the Grüneisen parameter. It holds for any frequency, hence it may be written for the Debye frequency \(\nu_D\)

\[ d \ln \nu_D / d \ln V = -\gamma \]

and since \(\nu_D\) and \(\theta_D\) are proportional, it follows

\[ d \ln \theta_D / d \ln V = -\gamma \quad \text{or} \quad d \ln \theta_D / dV = -\gamma / V. \]

Accordingly, we obtain for \(P_{vib}\)

\[ P_{vib} = \frac{9}{8} \left[ \frac{\gamma(V)}{V} \right] N k \theta_D + 3 \left[ \frac{\gamma(V)}{V} \right] N k T D(\theta_D/T) \]

or

\[ P_{vib} = \left[ \frac{\gamma(V)}{V} \right] E_{vib}, \]

which in terms of the assumed approximation \(\gamma/V = \gamma_0/V_0 = const.\) takes the form

\[ P_{vib} = \left( \gamma_0 / V_0 \right) E_{vib}. \]

These results render it possible to write Eqs.(1.7) in an explicit form:

\[ F(T, \varepsilon) = \sum_i E_{0i} \varepsilon^i + \frac{9}{8} N k \theta_D + N k T \left\{ 3 \ln \left[ 1 - \exp \left( -\theta_D/T \right) \right] - D(\theta_D/T) \right\}, \]
\[ E(T, \varepsilon) = \sum_i E_{0i} \varepsilon^i + \frac{9}{8} N k \theta_D + 3 N k T D(\theta_D/T), \]
\[ P(T, \varepsilon) = \sum_i P_{0i} \varepsilon^i + \frac{9}{8} (\gamma_0/V_0) N k \theta_D + 3 (\gamma_0/V_0) N k T D(\theta_D/T), \]

where the coefficients \(P_{0i}\) are given by Eq.(2.20).
These are the complete, the caloric and the thermal EOS for solids. They are defined over domains of the \((P,\varepsilon)\) and \((E,\varepsilon)\) planes formed by the set of experimental values of \(\varepsilon\), \(P\) and \(E\).

The integration constant \(E_00\) can be easily obtained from the caloric EOS. Let us write it in explicit form:

\[
E(T,\varepsilon) = E_{00} + E_{01}\varepsilon + E_{02}\varepsilon^2 + \ldots + \frac{9}{8}Nk\theta_D + 3NkTD(\theta_D/T).
\]

Since ambient conditions are assumed as a reference state, e.g. \(E(T=300K,\varepsilon=0) = 0\), we have

\[
E_{00} = -\frac{9}{8}Nk\theta_D - 900NkD(\theta_D/300).
\]

Knowing the complete EOS, we are able in principle to derive all the thermodynamic properties of the system under consideration by simple differentiation.

To calculate the temperature on the shock Hugoniot we may use either the caloric or the thermal EOS. Let us write the thermal EOS in the form \(T = f(T)\) and replace \(P(T,\varepsilon)\) with \(P_H(\varepsilon)\). We obtain

\[
T_H(\varepsilon) = \left(\frac{V_0}{3\gamma_0}\right) \left[ P_H(\varepsilon) - \sum_i \left\{ \left(\frac{i+1}{V_0}\right) E_{0i+1} - \frac{9}{8}(\gamma_0/V_0)Nk\theta_D \right\} \right] /NkD(\theta_D/T_H).
\]

This equation may be solved either graphically or by the method of successive approximations.

The simultaneous solution of the caloric and the thermal EOS in order to eliminate \(T\) defines the incomplete EOS

\[
P(E,\varepsilon) = (\gamma_0/V_0)E + \sum_i \{(i+1)/V_0\} E_{0i+1} - (\gamma_0/V_0)E_{0i}\varepsilon^i.
\]

This is the constitutive relation necessary to supplement the system of equations of fluid dynamics describing adiabatic compressible inviscid flow.

4. Conclusions

The assumptions that the free energy of a solid may be represented as a superposition of terms, corresponding to the cold compressed state, the quantum zero vibrations and the thermal vibrations of the crystal lattice, that the ratio \(\gamma(V)/V\) remains constant, and the Debye model for the specific heat, along with the experimental shock Hugoniot allow the derivation of its thermodynamic properties in explicit form. In the beginning a differential equation for the zero Kelvin isotherm is derived using the Mie-Grüneisen equation and the shock Hugoniot as a reference curve. An analytic solution to this equation is obtained in the form of power series. The arbitrary constant is expressed in terms of the specimen’s parameters at ambient conditions. Based on this solution, the complete, the caloric, the thermal and the incomplete EOS are obtained.

The proposed EOS are valid in the range of pressures and temperatures where the behaviour of the solid state is determined by the lattice thermal vibrations. At very
low and high temperatures the conduction electrons contribution must be taken into account. At higher temperatures the anharmonicity effects should be included as well.

The complete EOS is derived mainly within the framework of thermodynamics and it cannot predict phase transitions. Should a phase transition (or melting) be registered experimentally, the high-pressure phase (the melt) might be described using the technique, given in \[4\].

The obtained incomplete EOS derives from a thermodynamically consistent complete EOS and, therefore, it might serve as a reliable physical constitutive relation, required to elucidate how the characteristics of the material affect the propagation of shock waves. This is one of the aims of the present work.

Also, an equation for the shock temperature along the Hugoniot curve is derived.

Without the use of the algebraic programming system REDUCE \[8\] many of these expressions could not have been obtained easily.

In a forthcoming paper the proposed EOS will be applied to some solids for which the necessary shock-compression data are available.

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