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EQUATION OF STATE AND SHOCK ENTROPY OF CaCO_3

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EQUATION OF STATE AND SHOCK ENTROPY OF CaCO_3

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ABSTRACT

The energy released in a nuclear detonation can vaporize the surrounding medium. If one knows the gaseous^{*} equation of state of the medium, it can be used along with estimates of the shock entropy to predict such things as cavity size and seismic effects. We have calculated the equation of state and shock entropy for calcite (CaCO_3). We formulated the gaseous equation of state for pressures between 1 and 10^6 bar and temperatures from ambient to 1000 eV using a combination of the TIGER and IEEOS codes.

In this report, we also describe the calculational methods and the material parameters used to estimate shock entropy on the loading path. We show how these results can be used to estimate final states following unloading from a given point on the shock Hugoniot. A specific example, dense calcite at 900 kbar, is used to illustrate the method.

INTRODUCTION

The calculation of the effects of underground nuclear explosions on the surrounding medium is an area of active study at the Lawrence Livermore Laboratory. The energy released in a nuclear detonation is sufficient to vaporize the surrounding medium to such an extent that it becomes the principal constituent of the working gas. Thus, if one is to predict such things as cavity size, extent of cratering, and seismic effects, it is necessary to know the gaseous equation of state of the medium. The equation of state can be used with estimates of shock entropy to calculate the state of the medium following unloading from the shocked state.

Butkovich¹ developed gas equations of state for H_2O , NaCl , and silicate rocks. In this report, we describe the development of an equation of state for calcite (CaCO_3). We used Walsh and Christian's methods² to calculate the entropy for points along the shock Hugoniot in order to determine the isentropic unloading path.

^{*}We have used the term gaseous to include mixtures of gases and condensed species both liquid and solid.

MASTER

EQUATION OF STATE OF CaCO_3

Method

Butkovich¹ used LLL's Ionization Equation of State (IEEOS)³⁻⁷ to calculate equilibrium points for temperatures above 1 eV. For temperatures below 1 eV Butkovich used the HUG⁸ code to calculate equilibrium points for mixtures of the chemical species present. The HUG code treats the gas phase as an ideal-gas mixture and handles only one condensed phase.

We used the IEEOS code for the high-temperature region and the TIGER⁹ code for the thermochemical region. The TIGER code has the advantage that it can handle some aspects of non-ideal gas behavior and more than one condensed phase. As input data, TIGER requires covolumes of gas species for the Kistiakowsky-Wilson equation of state, enthalpies of formation of compounds, absolute entropy at 298 K, and the heat capacity at constant pressure as a function of temperature for all species. The IEEOS code requires all of the ionization potentials for each element.

Results

The results of the equilibrium calculations carried out with the TIGER and IEEOS codes are presented in Fig. 1, where energy is plotted as a function of pressure for constant temperature and constant density. The energy values are relative to CaCO_3 at 298 K. Since the IEEOS code reports energy values in relation to the atoms at 0 K, the values generated by the IEEOS code were adjusted to be relative to CaCO_3 at 298 K. The isodensity lines calculated for the thermochemical region fair smoothly into those calculated for the high-temperature region at pressures below 5×10^{-4} Mbar. Above this pressure, the 1 eV isotherm calculated by the IEEOS code was omitted and the low-temperature isodensity lines are faired into those calculated at 2 eV and higher. The TIGER code is preferred at 1 eV and high pressures because of the high stability of the molecular species CO and CO_2 at these conditions.

Some qualitative features of the system are apparent by following the 4000-K isotherm from low to high pressure (see Fig. 2). The first cusp observed with increasing pressure indicates the condensation of liquid CaO. At this temperature the system is completely vaporized at pressures below

Fig. 1. Equilibrium equation of state for CaCO_3 .

Fig. 2. The 4000-K isotherm from the equation of state of CaCO_3 .

3 bar. At 100 bar, complete vaporization occurs between 5000 and 6000 K. At higher pressures a second discontinuity is observed indicating the formation of condensed CaCO_3 .

In the energy-pressure plane, these discontinuities can be used to separate regions of complete vaporization, mixed gas and condensed phases, and complete condensation.

In Fig. 3 lines of constant entropy are shown as a function of the pressure and energy of the system. The entropy values are those calculated by TIGER and are absolute (i.e., relative to 0 K). The absolute entropy of CaCO_3 at 298 K is 0.221 cal/g·K.

Comparison With Ideal-Gas Calculations

Some of the results generated by the TIGER code were compared with ideal-gas calculations. This was done for several pressures along the 5000 K isotherm. For this purpose the following chemical equilibria were used:

Equilibrium constants for these reactions were calculated using the free-energy tabulations in the Shick¹⁰ and JANAF tables.¹¹ These four equations along with the relative proportions of atoms determined by the stoichiometry of CaCO_3 are sufficient to calculate the relative concentrations of the seven species. The results of

Table 1. Comparison of specific volumes at 5000 K calculated by TIGER code and ideal-gas law.

	Specific volume, cm ³ /g	
	5000 atm	10 000 atm
TIGER	1.89	1.24
Ideal gas	1.53	0.67

Fig. 3. Lines of constant entropy (isentropes) calculated by TIGER code.

the two calculations are compared in Fig. 4 and Table 1. At high pressures the TIGER code calculates considerably larger specific volumes (lower density) for the system than does the ideal-gas law. This is due to intermolecular repulsion, which becomes increasingly important at high pressure. Figure 4 shows that the two calculations are in close agreement up to pressures of 1000 atm.

CALCULATION OF SHOCK ENTROPY

For underground nuclear explosions, the formation of the working gas is due primarily to shock vaporization of the surrounding medium. Therefore, we would like to be able to calculate the unloading path of the material after the shock wave passes and to determine the final state of the material as a function of pressure.

Method

Our approach was to estimate the entropy for points along the shock Hugoniot. In this way the equation of state isentropes could be connected to the shock Hugoniot. The first step was to calculate ΔS for the shock process.

Shock entropy can be calculated as a two-step process (see Fig. 5). For the constant-volume heating step:

$$\left(\frac{\partial S}{\partial T}\right)_V = C_v/T$$

$$\Delta S = C_v \ln T_1/T_0 . \quad (1)$$

For the constant-temperature compression step:

$$\left(\frac{\partial S}{\partial V}\right)_T = \left(\frac{\partial P}{\partial T}\right)_V = \frac{\alpha}{\beta}$$

$$\Delta S = \frac{\alpha}{\beta} (V_f - V_0) . \quad (2)$$

Fig. 4. Comparison of concentrations of species as calculated from TIGER code and ideal-gas laws at 5000 K.

Fig. 5. Steps used in calculating the shock entropy.

where,

α = coefficient of thermal expansion = $1/V(\partial V/\partial T)_P$

β = coefficient of isothermal compressibility = $1/V(\partial V/\partial P)_T$.

We use the method of Walsh and Christian^{2,12} to calculate T_1 for points along the shock Hugoniot. Several methods utilizing experimental shock data and ambient materials properties will be used to evaluate Eqs. (1) and (2).

Walsh-Christian Equation

The equation derived by Walsh and Christian for calculating temperatures along a Hugoniot is,

$$T_1(V_1) = T_0 \exp\left[\frac{\Gamma}{V} (V_0 - V_1)\right] + \frac{\exp\left[-\left(\frac{\Gamma}{V}\right)V_1\right]}{C_v} \int_{V_0}^{V_1} \left[f(V_1) e^{\frac{\Gamma}{V} V_1} dV \right], \quad (3)$$

where

$$f(V_1) = \frac{1}{2} \left[\frac{dP_1}{dV_1} (V_0 - V_1) + P_1 \right] \quad (4)$$

$$\Gamma = \frac{\alpha V}{\beta C_v} = \text{Grüneisen parameter} \quad (5)$$

and

V = specific volume

C_v = heat capacity at constant volume.

Derivation and Assumptions

Walsh and Christian derived Eq. (3) by combining the thermodynamic identity,

$$TdS = C_v dT + \left(\frac{\partial P}{\partial T} \right)_V TdV, \quad (6)$$

with the first law of thermodynamics,

$$dE = TdS - PdV \quad (7)$$

and the equation for shock energy

$$E_1 - E_0 = \frac{1}{2} (P_0 + P_1)(V_0 - V_1) \quad (8)$$

Essential to the derivation are assumptions that C_v and α/β are constant throughout the shock process. The assumption that Γ/V is a constant is an approximation of the results reported in Ref. 13.

There is some uncertainty as to the correct value of C_v to use in the calculation of Eq. (3). At room temperature, C_v for CaCO_3 is 0.194 cal/g·K. The Dulong-Petit value, which is the approximate high temperature limit, is 0.298 cal/g·K. Data, given in Ref. 14, indicate a value of 0.317 cal/g·K for the heat capacity at constant pressure, at 1400 K.

Solution

Solution of the term $f(V_1)$ in Eq. (3) depends solely on the pressure-volume relationship of the experimental Hugoniot. The definite integral of Eq. (3) must be evaluated by numerical or graphical methods. To evaluate $f(V_1)$ we need an analytic expression for P as a function of V . For this purpose we use (see Refs. 15 and 16)

$$P_1 - P_0 = \frac{a_0^2(1 - v_1/v_0)}{v_0[1 - b(1 - v_1/v_0)]^2}, \quad (9)$$

where

V_0 = specific volume at zero pressure

b = slope of the shock velocity vs particle velocity relationship

a_0 = shock velocity at zero particle velocity (the hydrodynamic speed of sound in the solid).

Equation (9) is derived from the conservation laws for mass and momentum across a shock wave and the assumption of a linear relationship between shock velocity, U_S , and particle velocity U_p :

$$U_S = a_0 + bU_p \quad (10)$$

Differentiating P with respect to V in Eq. (9) gives

$$\frac{dP_1}{dV_1} = -a_0^2 [V_0 - b(V_0 - V_1)]^{-2} - 2ba_0^2 [V_0 - V_1] [V_0 - b(V_0 - V_1)]^{-3} \quad (11)$$

for the slope of the shock Hugoniot.

Values of dP_1/dV_1 determined by Eq. (11) can be used in Eqs. (3) and (4) to calculate T_1 .

Dense Calcite

The following values for materials properties were used:

$V_0 = 0.37 \text{ cm}^3/\text{g}$, which corresponds to $\rho = 2.703 \text{ g/cm}^3$. The theoretical density of calcite is 2.711 g/cm^3

$b = 1.484$

$a_0 = 3.675 \times 10^5 \text{ cm/s}$.

Values of b and a_0 were determined from the experimental shock Hugoniot for dense calcite. This plot, based on data for limestone and marble,¹⁷ is shown in Fig. 6. The data for shock velocity versus particle velocity are plotted in Fig. 7.

This treatment of Hugoniot data is idealized in that zero initial porosity is assumed and phase transitions and discontinuities along the Hugoniot have been neglected. This is probably not a true picture.¹⁸⁻²¹ Nevertheless, when the Hugoniot data are plotted as pressure vs volume, Eq. (9) fits the data well (see Fig. 6).

Grüneisen Parameter

There are several ways to estimate (Γ/V) : from thermodynamic data for calcite, from the Hugoniot shock data, and from an equation of state proposed by Cook and Rogers.²²

Fig. 6. Shock Hugoniot for dense calcite to 900 kbar. The data points are for marble and limestone. The curve is a plot of Eq. (9).

Fig. 7. Shock velocity vs particle velocity for calcite.

Table 2. Thermodynamic properties of CaCO_3 (taken from Ref. 23).

$$\alpha_V = 13.1 \times 10^{-6} \text{ K}^{-1}$$

$$\beta = 1.4 \times 10^{-6} \text{ bar}^{-1}$$

$$C_V = 0.194 \text{ cal/g}\cdot\text{K}$$

$$V = 0.37 \text{ cm}^3/\text{g}$$

$$\Gamma = \frac{\alpha V}{\beta C_V} = 0.427$$

Thermodynamic Data — Table 2

gives the properties of CaCO_3 (calcite).

The quantity (Γ/V) , calculated from thermodynamic properties, is therefore $0.427/0.37 = 1.153$.

Hugoniot Shock Data — The

Grüneisen parameter can also be calculated from the slope of the shock velocity vs particle velocity curve, b. At limiting shock conditions¹⁶ ($U_p \rightarrow \infty$)

$$\Gamma = 2(b - 1) \quad (12)$$

Taking $b = 1.484$

$$\Gamma = 0.968.$$

Also at limiting shock conditions¹⁶

$$\left(\frac{V_1}{V_0}\right)_L = 1 - 1/b \quad (13)$$

or $V_1 = 0.1207 \text{ cm}^3/\text{g}$ and $(\Gamma/V) = 0.968/0.1207 = 8.02$.

This estimate is almost certainly too high as it is unlikely that the limiting slope of the U_s vs U_p curve is 1.484. For most substances the linear relationship shown in Fig. 7 is not maintained at very high pressures and the slope decreases.

At zero shock ($U_p \rightarrow 0$), according to Wentorf²⁴

$$\Gamma = 2b - 1 \quad (14)$$

or $\Gamma = 1.968$ and with $V = 0.37$, $(\Gamma/V) = 5.319$.

The Grüneisen constant can also be calculated by Eq. (5) with a value for isothermal compressibility, β , obtained from the speed of sound a_0

$$\frac{1}{\rho a_0^2} = \beta_s \approx \beta_T, \quad (15)$$

where β_s is the isentropic compressibility, which is close to the isothermal compressibility, β_T , if C_v/C_p is close to unity.

From Eq. (15)

$$\beta_T = 2.74 \cdot 10^{-6} \cdot \text{bar}^{-1}.$$

This value is nearly twice that reported in Ref. 23.

From Eq. (5)

$$\Gamma = 0.218$$

$$\frac{\Gamma}{V} = \frac{0.218}{0.37} = 0.589.$$

Equation of State — Another estimate of the Grüneisen constant can be obtained from an equation of state proposed by Cook and Rogers^{22,25,26}

$$\frac{V}{V_0} = \left(1 + \frac{P}{P_i}\right)^{-\frac{1}{1+\Gamma}}, \quad (16)$$

where P_i is a generalized internal pressure and is approximated by the cohesive energy density:

$$P_i = \epsilon_0/V_0, \quad (17)$$

and ϵ_0 is the cohesive energy density at absolute zero. Equation (16) gives a very close fit to the experimental shock Hugoniot in Fig. 6 if $P_i = 71$ kbar and $\Gamma = 4.09$. This is the largest value of Γ we will use, and

$$\frac{\Gamma}{V} = \frac{4.09}{0.37} = 11.05$$

Shock Entropy

To illustrate the problem with a specific example, we calculated the shock entropy for calcite at 900 kbar using several values of the Gruneisen parameter and two values for C_v . In each case the ratio α/β used in the calculation of the constant temperature step was chosen to satisfy Eq. (5).

The results are given in Table 3, and plotted in Fig. 8 for three values of Γ/V . Also plotted in Fig. 8 is an entropy calculation using an approximation which is valid for weak shock waves²⁴:

$$s_1 - s_0 = - \frac{1}{12T_0} \left(\frac{\partial^2 P}{\partial v^2} \right)_{s_0} (v_1 - v_0)^3. \quad (18)$$

(The second derivative at the origin in Eq. (18) is obtained by taking the derivative of Eq. (11) and setting $v_1 = v_0$.)

Table 3. Shock entropy for dense calcite at 900 kbar.

$\Gamma/V,$ cm^3/g	$\Delta S, \text{ cal/g}\cdot\text{K}$	
	$C_v = 0.194$ cal/g·K	$C_v = 0.298$ cal/g·K
0	0.518	0.679
0.589	0.504	0.657
1.153	0.493	0.642
5.32	0.398	0.504
8.02	0.343	0.425
11.05	0.288	0.349

The most reasonable values for (Γ/V) are 5.32 and 8.02. Within this range the calculated values for shock entropy given in Table 3 indicate only a slight dependence of ΔS on (Γ/V) and C_v . Nevertheless, as will be shown later, small entropy changes correspond to quite large differences in the E-P plane.

Porous Calcite

We also did the same calculation for porous calcite. Figures 9 and 10 present shock Hugoniot data

Fig. 8. Shock entropy plotted as a function of pressure. The various curves show the effect of variations of C_V and Γ/V on this relationship. There is also a calculation made with a weak shock approximation.

Fig. 9. Shock velocity vs particle velocity for porous CaCO_3 .

Fig. 10. Shock Hugoniot for porous CaCO_3 . The shock Hugoniot for dense CaCO_3 is included for comparison.

for porous calcite ($V_0 = 0.574 \text{ cm}^3/\text{g}$) taken from van Thiel's¹⁷ compilation. The data are scattered, and the choice of a_0 and b for use in Eq. (9) is somewhat arbitrary. One constraint applied in choosing these parameters was that the Hugoniot for porous CaCO_3 generated by Eq. (9) not cross the Hugoniot for dense CaCO_3 when extrapolated to 900 kbar. (Figure 10 also includes the Hugoniot for dense CaCO_3 for comparison.)

As before, values of (Γ/V) from limiting shock and zero shock conditions can be generated by Eqs. (12), (13) (limiting shock), and (14) (zero shock). (No attempt was made to calculate a value of (Γ/V) from the thermodynamic properties since this would require an estimate of α and β for the porous material.) The values are,

$$\left(\frac{\Gamma}{V}\right) = 2.65 \text{ g/cm}^3 \quad (\text{zero-shock conditions})$$

and

$$\left(\frac{\Gamma}{V}\right) = 4.39 \text{ g/cm}^3 \quad (\text{limiting-shock conditions}).$$

The procedure described earlier was used to estimate shock entropy for porous CaCO_3 . The results of the calculations are plotted in Fig. 11 along with the results of the entropy calculation for dense CaCO_3 for comparison.

Some conclusions can be drawn by examination of Fig. 11:

- a) The shock entropy is greater for porous CaCO_3 than for dense CaCO_3 at equivalent pressures.
- b) The (Γ/V) parameter derived from the zero-shock condition leads to higher estimates of shock entropy than the parameter at limiting conditions.
- c) Considering only the calculations based on zero-shock and limiting-shock estimates of (Γ/V) (i.e., neglecting the thermodynamic estimate of (Γ/V) and the weak shock approximation),

ΔS for dense CaCO_3 is 0.34 to 0.50 cal/g·K
at 900 kbar

and ΔS for porous CaCO_3 is 0.49 to 0.78 cal/g·K.

Fig. 11. Shock entropies vs pressure for CaCO_3 .

DISCUSSION OF RESULTS

In principle, the combination of estimates of shock entropy for dense CaCO_3 and for porous CaCO_3 plus the equation of state, which locates isentropes in the energy-pressure plane, can be used to calculate the state of material following unloading from a shocked state. In practice, there is an uncertainty in the estimate of shock entropy due to the difficulty of selecting the proper value of the Grüneisen constant, Γ , and the questionable validity of the assumption that (Γ/V) is constant throughout the shock process. This uncertainty leads to an uncertainty in locating the final unloaded state.

For example, assume that for both porous and dense CaCO_3 , the correct values of (Γ/V) probably lie between the zero-shock and limiting-shock assumptions. The calculated shock entropy at 900 kbar for dense CaCO_3 will then lie between 0.34 and 0.50 cal/g·K. The calculated shock entropy for porous CaCO_3 at 900 kbar will lie between 0.49 and 0.78 cal/g·K. Adding 0.22 cal/g, which is the entropy at 298 K, the unloading path from 900 kbar for dense CaCO_3 will be along a path between the 0.56 and 0.72 isentropes. The path for porous CaCO_3 will be between the 0.71 and 1.0 isentropes. For dense CaCO_3 the system energy at 100 atm will be below 400 cal/g, possibly in a region of partial disassociation to CaO and CO_2 . For porous calcite the energy at 100 atm will lie between 400 and 850 cal/g ranging from partial to complete dissociation to CaO and CO_2 . In neither case will CaO be vaporized. That is, neither dense CaCO_3 or the porous CaCO_3 examined here can be completely vaporized by unloading from a 900-kbar shock.

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