On the Calculation of the Seismic Parameter ϕ at High Pressure and High Temperatures

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Comparison of the Murnaghan equation of state with the Birch equation shows that, for a given value of pressure, the values of (ρ/ρ_0) calculated from the two equations differ by less than 1% to a pressure equal to 0.5 K_0 (where K_0 is the zero-pressure isothermal bulk modulus), but the corresponding values of the seismic parameter ϕ differ by 10%. The value of ϕ is extremely sensitive to the choice of the equation of state because ϕ is the derivative of pressure with respect to density. The good agreement between the two equations of state for pressure as a function of density observed for some materials does not imply the same agreement in the relationship between ϕ and pressure. Expressions for $\phi(P)$ that take into account the first order nonlinear dependence of the bulk modulus on pressure are presented, and their applications are discussed. Temperature correction of the pressure-dependent ϕ is also considered.

Comparison of the seismic parameter ϕ_{LAB}, determined in the laboratory for various materials, with the values actually observed in the field, ϕ_{FLD}, can be used to estimate the composition of a homogeneous isothermal layer within the earth. If a particular equation of state is assumed, then the seismic parameter may be written as a function of pressure because the definition of the adiabatic bulk modulus K_s implies that

$$\phi = \left(\frac{\partial P}{\partial \rho}\right)_s \tag{1}$$

Birch [1939] used the Murnaghan theory of finite strain to calculate the rate of change of seismic velocities with pressure. O. L. Anderson presented an equation for a pressure-dependent ϕ based on the Murnaghan equation of state and illustrated its applicability at high pressure. He concluded [*O. L. Anderson*, 1966, p. 730] that 'Birch's equation of state, in its form which is appropriate to a general value of K_0, leads to essentially the same results as does the Murnaghan equation'; we believe the two equations lead to different results.

In this paper we compare the values calculated for ϕ from both the Murnaghan and the Birch equations of state and discuss the sensi-

tivity of $\phi(P)$ to the choice of the equation of state; we believe the Birch form superior to that of Murnaghan. Expressions for $\phi(P)$ that take into account the first-order nonlinear dependence of the bulk modulus on pressure are given, and their implications are discussed. Correction of the pressure-dependent ϕ for temperature is considered.

SENSITIVITY OF $\phi(P)$ TO THE CHOICE OF EQUATION OF STATE

The equations of state most widely used in geophysics are those of *Murnaghan* [1944, 1949] and *Birch* [1939, 1947, 1952]. We examine the dependence of $\phi(P)$ on the form of the equation of state used to describe the elastic behavior of solids.

The Murnaghan equation of state is derived from the assumption that bulk modulus is a linear function of pressure: $K(P) = K_0 + mP$, where K_0 is the adiabatic bulk modulus evaluated at zero pressure, and m is a material constant defined by $m = \{(\partial K/\partial P)_s\}_{P=0}$. Since $K = \rho(dP/d\rho)$,

$$P_M = (K_0/m)[(\rho/\rho_0)^m - 1] \tag{2}$$

The subscript M denotes parameters calculated from the Murnaghan equation of state.

The Birch equation of state, derived from Murnaghan's theory of finite strain [*Murnag-*

han, 1951] with cubic and quadratic terms of strain retained in the Helmholtz free energy [*Birch*, 1947, 1952], leads to

$$P_B = (3K_0/2)[(\rho/\rho_0)^{7/3} - (\rho/\rho_0)^{5/3}]$$

$$\cdot\{1 + (\tfrac{3}{4})(m - 4)[(\rho/\rho_0)^{2/3} - 1]\} \qquad (3)$$

The subscript B refers to the parameters calculated from the Birch equation of state.

From equation 2, we find the derivative

$$dP_M/d\rho = \phi_0(\rho/\rho_0)^{m-1} \qquad (4)$$

where $\phi_0 = (K_0/\rho_0)$. To express ϕ as a function of pressure, we substitute equation 2 in the form

$$\rho/\rho_0 = [1 + m(P_M/K_0)]^{1/m} \qquad (5)$$

and obtain

$$\phi_M = dP_M/d\rho$$

$$= (K_0/\rho_0)[1 + m(P_M/K_0)]^{(m-1)/m} \qquad (6)$$

Equation 6 corresponds to equation 8 of *O. L. Anderson's* [1966] paper, and it is noted that he derived this expression in a different way.

Similarly, from the Birch equation of state, we have

$$P_B = (3K_0/2)y^5\{(y^2 - 1) + b_1(y^2 - 1)^2\} \qquad (7)$$

and

$$\phi_B = dP_B/d\rho = (\phi_0/3)\{3y^4[1 + 2b_1(y^2 - 1)] + (5/y^3)(P_B/K_0)\} \qquad (8)$$

where $y = (\rho/\rho_0)^{1/3}$ and $b_1 = (3/4)(m - 4)$. To obtain ϕ_B as a function of pressure, the Birch equation of state must be solved numerically for (ρ/ρ_0) as a function of pressure.

It has previously been recognized that the Murnaghan equation 2 will be limited to values of $P < 0.5K_0$ in estimating (V/V_0) [e.g., *O. L. Anderson*, 1968, p. 170]. We show below that its validity for ϕ does not extend as high as $P \simeq 0.5K_0$.
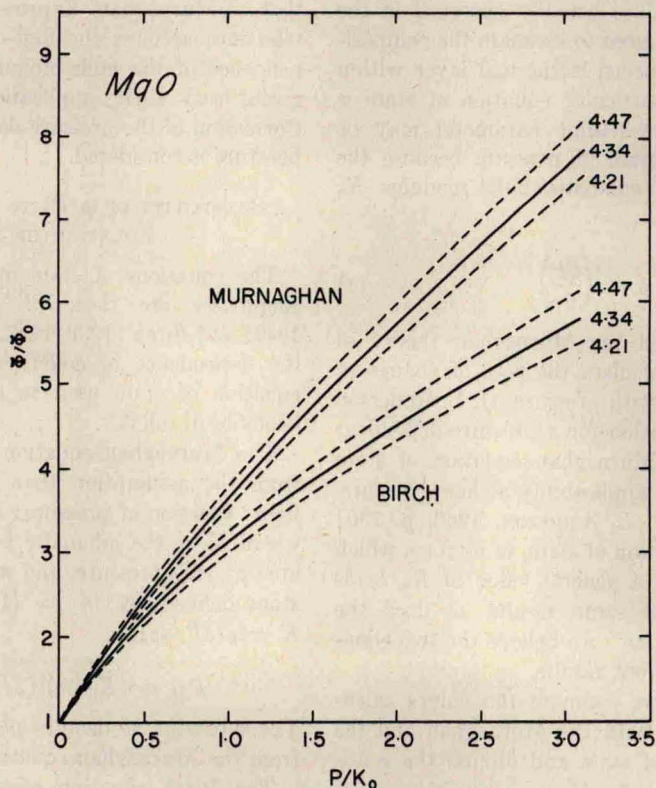

Fig. 1. Comparison of the seismic ϕ calculated from the Birch and the Murnaghan equations for periclase (at 298°K).

Numerical results for $m = 4$, a common value, are listed in the appendix of this paper in normalized form. For a given value of (P/K_0), the computed values of (V/V_0) and (ϕ/ϕ_0) from the Murnaghan and the Birch equations are tabulated. Note that because K_0 and K_0' are adiabatic, the values of (ϕ/ϕ_0) are also adiabatic. Differences between $(\phi/\phi_0)_B$ and $(\phi/\phi_0)_M$ are small at low values of P/K_0 but are larger at high values of P/K_0; differences between $(V/V_0)_B$ and $(V/V_0)_M$ are small at all values of P/K_0 less than one. Although the value of $(V/V_0)_B$ differs from the corresponding $(V/V_0)_M$ by less than 1% at a pressure corresponding to 0.5 K_0, the value of $(\phi/\phi_0)_B$ differs from $(\phi/\phi_0)_M$ by 10%. At pressure in the vicinity of the bulk modulus of a solid, the difference between $(V/V_0)_B$ and $(V/V_0)_M$ is only 2½%, but the corresponding difference for (ϕ/ϕ_0) is at least 17%. For other values of m, these differences in (V/V_0) and (ϕ/ϕ_0) resulting from the Murnaghan and the Birch equations are tabulated in the appendix as a function of pressure.

The sensitivity of the seismic $\phi(P)$ to the form of the equation of state is apparent. For an equation of state to provide as precise a formula for the seismic ϕ as a function of pressure as it does for pressure as a function of density, the equation of state must not only fit the experimental pressure-density curves sufficiently well, but it must also have the correct *functional form* so that the derivatives match equally well. Comparison of an equation of state with experimental data on compression, as frequently seen in the literature, does not provide a good test of the validity of the expression for $\phi(P)$ where ϕ is derived from the equation of state.

The Murnaghan and Birch equations for $\phi(P)$ may be illustrated with periclase (MgO). All the experimental data necessary to find K_0 and m in equations 6 and 8 are well established [e.g., *O. L. Anderson et al.*, 1968], and data on shock-wave compression to about 2.6 mb are available [*Al'tshuler et al.*, 1965; *McQueen and Marsh*, 1966] to test the extrapolations. Periclase is interesting to geophysics because it is a rock-forming mineral and also because it has been proposed as a separate phase in the lower mantle. Note that both the Murnaghan and the Birch equations for $\phi(P)$ are completely speci-

fied by the values of K_0 and m. Although these quantities are readily measurable with several different methods,[1] the ultrasonic measurements of compressional and shear velocities as a function of pressure result in the most accurate values of K_0 and m [e.g., *Daniels and Smith*, 1963; *O. L. Anderson*, 1965]. For the initial parameters we used $K_s = 1623$ kb and $(\partial K_s/\partial P)_s = 4.34$ (both evaluated at $P = 0$ and $T = 300°K$) [*Chang and Barsch*, 1969; *Chung and Simmons*, 1969], with the result that the adiabatic $\phi_0 = 45.3$ km/sec². Using these values in equations 6 and 8, (ϕ/ϕ_0) as a function of pressure was calculated; the results are shown in Figure 1. Although $(\phi/\phi_0)_M$ is indistinguishable from $(\phi/\phi_0)_B$ at pressures below 0.05 K_0, the two parameters are very different at pressures greater than 0.05 K_0. For example, at 1.4 mb (the pressure corresponding to the core-mantle boundary), the value calculated from the Murnaghan equation is 18% larger than that calculated from the Birch equation, even though the density difference is only about 2%, as seen in Figure 2.[2]

Errors in K_0 and m affect the precision of $\phi(P)$. Provided ultrasonic measurements are appropriately made, the value of K_0 can be determined to an accuracy of a few parts in 10^4, and the effects of this magnitude on $\phi(P)$ is small. An error in m frequently amounts to as much as 3% in the usual ultrasonic measurements. The effects of a 3% error in m are

[1] One of the earlier methods is an isothermal compression measurement of volume (or length) typified by work of *Bridgman* [1949]. An X-ray diffraction method, in which a change in dimension of the unit cell is measured as a function of pressure, has been used by a number of investigators [e.g., *Drickamer et al.*, 1966; *McWhan*, 1967]. Shock-wave compression such as the work of McQueen and colleagues [*McQueen et al.*, 1967] has been used to estimate K_0 and m [see, for example, *D. L. Anderson and Kanamori*, 1968]. The ultrasonic methods pioneered by *Lazarus* [1949] have been improved to a degree that their data yield estimates of K_0 to four significant figures and m to three.

[2] The earlier correlation of the ultrasonic and shock-wave data established for periclase [*O. L. Anderson*, 1965, 1966] appears to be fortuitous since the ultrasonic K_0 of this material was too *high* (compare the former value of 1717 kb with a revised value of 1622 kb) and ultrasonic m was too *low* (compare 3.96 against a new value 4.55; see *O. L. Anderson et al.* [1968]).
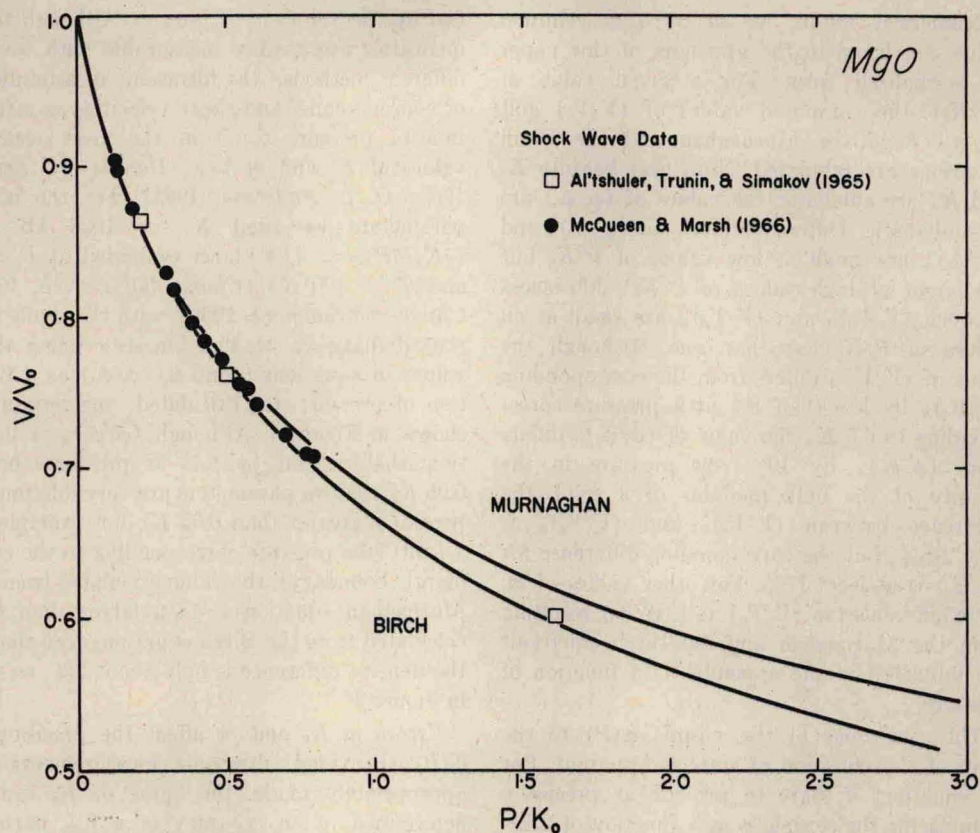
Fig. 2. Comparison of the calculated pressure-volume relations based on the Birch and the Murnaghan equations of state with shock-wave compression data for periclase. The equation-of-state parameters used are the adiabatic values evaluated at 298°K and zero pressure.

illustrated in Figure 1. Note that, at 1.4 mb, uncertainties seen in the seismic ϕ values resulting from the Murnaghan and the Birch equations are about 4% each. It seems, then, that the accuracy of the calculation of ϕ at high pressure is limited mainly by the accuracy with which m can be determined from ultrasonics.

MURNAGHAN EQUATION VERSUS BIRCH EQUATION

The general superiority of the Birch equation of state over that of Murnaghan will be discussed elsewhere with respect to the pressure-volume relation of various solids. Use of the Murnaghan equation of state leads to overestimates of the volume at high pressure. The reason here is associated not only with the assumption of constant m but also with an inadequacy of the functional form of the equa-

tion itself. Analysis of Bridgman's data [*Bridgman*, 1964] on the compression of various solids reveals a nonlinear behavior of the bulk modulus. *Chang and Barsch* [1967] observed ultrasonically a nonlinear pressure dependence of all second-order elastic constants for single crystal CsCl, CsBr, and CsI at pressures as low as 3 to 4 kb. The significance of their experimental finding is that deviation from constant m may amount to as much as 40 to 50% at pressures in the vicinity of the bulk modulus of solids and raises a question as to the general validity of the Murnaghan equation of state and the Murnaghan assumption.

Bullen [1947, 1949] discussed the nonlinear dependence of the bulk modulus with pressure in connection with the compressibility of the earth's interior. More recently, *Ruoff* [1967] expressed the experimental bulk modulus in

terms of a power series of P as

$$K(P) = -V\left(\frac{dP}{dV}\right)_s$$

$$= K_0 + K_0'P + \tfrac{1}{2}K_0''P^2 \qquad (9)$$

where $K_0'' = \{(\partial^2 K/\partial P^2)_s\}_{P=0}$ is the coefficient of the first-order nonlinear term in the bulk modulus.

Integration of equation 9 in the same manner as *Murnaghan* [1944] shows that

$$\ln\left[\frac{x_2}{x_1}\left(\frac{P - x_1}{P - x_2}\right)\right] = \ln\left(\frac{\rho}{\rho_0}\right)^{\xi} \qquad (10)$$

where

$$\xi = [(K_0')^2 - 2K_0K_0'']^{1/2} \qquad (11)$$

and

$$x_1, x_2 = -(K_0'/K_0'') \pm (\xi/K_0'') \qquad (12)$$

Rewriting equation 10, the 'second-order Murnaghan equation of state' is found as

$$P = [x_1(1 - Z^{\xi})]/[1 - (x_1/x_2)Z^{\xi}] \qquad (13)$$

where the coefficients ξ, x_1, and x_2 are given by equations 11 and 12 respectively, and $Z = (V_0/V) = (\rho/\rho_0)$.

Similarly, the 'second-order Birch equation of state' may be found as

$$P = (3K_0/2)y^5[(y^2 - 1)$$

$$+ b_1(y^2 - 1)^2 + b_2(y^2 - 1)^3] \qquad (14)$$

where

$$b_1 = 3/4(K_0' - 4) \qquad (15)$$

and

$$b_2 = 1/24[143 + 9(K_0' - 7)K_0'$$

$$+ 9K_0K_0''] \qquad (16)$$

and

$$y = (V_0/V)^{1/3} = (\rho/\rho_0)^{1/3}$$

as before.

Based on these second-order equations of state, the corresponding expressions for the $\phi(P)$ are:

$$\phi(P)_{\text{Murnaghan}} = \phi_0\left[1 + \left(\frac{K_0'}{K_0}\right)P\right.$$

$$+ \left(\frac{K_0''}{2K_0}\right)P^2\left]\left[\frac{x_1}{x_2}\left(\frac{P - x_2}{P - x_1}\right)\right]^{1/\xi} \qquad (17)$$

and

$$\phi(P)_{\text{Birch}} = \frac{\phi_0}{3}\{3y^4[1 + 2b_1(y^2 - 1)$$

$$+ 3b_2(y^2 - 1)^2 + \frac{5}{y^3}(P/K_0)]\} \qquad (18)$$

These last two equations are new and account for the first-order nonlinear behavior of the bulk modulus with pressure. Their usefulness arises from the fact that all the parameters can be evaluated from either the quantities determined from ultrasonic measurements at modest-pressure range or from low-pressure ultrasonic data combined with high-pressure compression data (after the appropriate thermodynamic transformation).

TABLE 1. Comparison of Volume and the Seismic Parameter ϕ as a Function of Pressure (for $m = 4$)

(P/K_0)	$(V/V_0)_B$	$(V/V_0)_M$	$(\phi/\phi_0)_B$	$(\phi/\phi_0)_M$
0.000	1.000	1.000	1.000	1.000
0.020	0.981	0.981	1.059	1.059
0.040	0.964	0.964	1.115	1.118
0.060	0.948	0.948	1.169	1.175
0.080	0.933	0.933	1.222	1.231
0.100	0.919	0.919	1.272	1.287
0.120	0.906	0.907	1.322	1.342
0.140	0.894	0.895	1.369	1.396
0.160	0.883	0.884	1.416	1.449
0.180	0.872	0.873	1.462	1.502
0.200	0.862	0.863	1.506	1.554
0.220	0.852	0.854	1.550	1.606
0.240	0.843	0.845	1.593	1.657
0.260	0.835	0.837	1.634	1.707
0.280	0.826	0.829	1.675	1.757
0.300	0.818	0.821	1.716	1.806
0.320	0.811	0.814	1.755	1.855
0.340	0.803	0.807	1.795	1.904
0.360	0.796	0.800	1.833	1.952
0.380	0.789	0.794	1.871	2.000
0.400	0.783	0.788	1.908	2.048
0.420	0.776	0.782	1.945	2.095
0.440	0.770	0.776	1.981	2.141
0.460	0.765	0.770	2.017	2.188
0.480	0.759	0.765	2.052	2.234
0.500	0.753	0.760	2.087	2.280
0.600	0.728	0.736	2.255	2.504
0.700	0.706	0.716	2.414	2.722
0.800	0.687	0.699	2.566	2.934
0.900	0.669	0.683	2.712	3.141
1.000	0.653	0.669	2.853	3.344
1.500	0.592	0.615	3.491	4.304
2.000	0.549	0.577	4.054	5.196
2.500	0.516	0.549	4.566	6.040
3.000	0.490	0.527	5.039	6.846

TABLE 2. Comparison of Volume and the Seismic Parameter ϕ as a Function of Pressure (for $m = 5$)

(P/K_0)	$(V/V_0)_B$	$(V/V_0)_M$	$(\phi/\phi_0)_B$	$(\phi/\phi_0)_M$
0.000	1.000	1.000	1.000	1.000
0.020	0.981	0.981	1.078	1.079
0.040	0.964	0.964	1.153	1.157
0.060	0.949	0.949	1.225	1.234
0.080	0.935	0.935	1.295	1.309
0.100	0.922	0.922	1.362	1.383
0.120	0.910	0.910	1.427	1.456
0.140	0.899	0.899	1.491	1.529
0.160	0.888	0.889	1.553	1.600
0.180	0.878	0.880	1.614	1.671
0.200	0.869	0.871	1.674	1.741
0.220	0.860	0.862	1.732	1.810
0.240	0.852	0.854	1,789	1.879
0.260	0.844	0.847	1.845	1.947
0.280	0.836	0.839	1.900	2.015
0.300	0.829	0.833	1.955	2.081
0.320	0.822	0.826	2.008	2.148
0.340	0.816	0.820	2.061	2.214
0.360	0.809	0.814	2.113	2.279
0.380	0.803	0.808	2.164	2.344
0.400	0.797	0.803	2.214	2.408
0.420	0.792	0.797	2.264	2.472
0.440	0.786	0.792	2.314	2.536
0.460	0.781	0.788	2.362	2.599
0.480	0.776	0.783	2.411	2.662
0.500	0.771	0.778	2.458	2.724
0.600	0.749	0.758	2.689	3.031
0.700	0.729	0.740	2.910	3.331
0.800	0.712	0.725	3.122	3.624
0.900	0.697	0.711	3.327	3.911
1.000	0.683	0.699	3.525	4.193
1.500	0.629	0.652	4.439	5.540
2.000	0.590	0.619	5.262	6.809
2.500	0.561	0.594	6.023	8.022
3.000	0.537	0.574	6.737	9.190

A form of the second-order Murnaghan equation of state, equation 13, has been presented earlier by *Ruoff* [1967] and by G. R. Barsch and Z. P. Chang ('Ultrasonic and static equation of state for cesium halides,' in *Accurate Characterization of the High-Pressure Environment*, unpublished manuscript, 1970). The equation of state given by Barsch and Chang is similar to equation 14 in this paper.

Crystalline solids undergoing compression without a phase change are characterized by $K(P)$ increasing monotonically with pressure and $K'(P)$ decreasing monotonically with pressure. The second condition requires that $K'' < 0$; such behavior was ultrasonically observed for three cesium halides [*Chang and Barsch*, 1967].

Because $K_0'' < 0$, equation 9 implies the existence of a finite pressure at which $K(P)$ becomes negative. Yet $K(P)$ cannot be negative, by the first law of thermodynamics. Use of the second-order Murnaghan equation of state to extrapolate density to high pressure leads to impossible results. Similarly, equation 17 should not be used to extrapolate the seismic $\phi(P)$. The Birch equation of state, which is a phenomenological equation based on a rapidly converging Taylor expansion of the interatomic potential, appears to describe experimental compression curves of solids more closely than any other equation of state yet known. For this reason, the ultrasonic method discussed here for calculating ϕ at high pressure is based on the use

TABLE 3. Comparison of Volume and the Seismic Parameter ϕ as a Function of Pressure (for $m = 6$)

(P/K_0)	$(V/V_0)_B$	$(V/V_0)_M$	$(\phi/\phi_0)_B$	$(\phi/\phi_0)_M$
0.000	1.000	1.000	1.000	1.000
0.020	0.981	0.981	1.097	1.099
0.040	0.965	0.965	1.189	1.196
0.060	0.950	0.950	1.278	1.292
0.080	0.936	0.937	1.363	1.386
0.100	0.924	0.925	1.445	1.479
0.120	0.913	0.914	1.525	1.571
0.140	0.902	0.903	1.602	1.662
0.160	0.892	0.894	1.677	1.752
0.180	0.883	0.885	1.751	1.841
0.200	0.875	0.877	1.823	1.929
0.220	0.866	0.869	1.893	2.016
0.240	0.859	0.862	1.962	2.103
0.260	0.851	0.855	2.029	2.189
0.280	0.844	0.848	2.096	2.274
0.300	0.838	0.842	2.161	2.358
0.320	0.831	0.836	2.225	2.442
0.340	0.825	0.831	2.289	2.526
0.360	0.819	0.826	2.351	2.609
0.380	0.814	0.820	2.413	2.691
0.400	0.808	0.815	2.473	2.773
0.420	0.803	0.811	2.533	2.854
0.440	0.798	0.806	2.592	2.935
0.460	0.793	0.802	2.651	3.015
0.480	0.789	0.798	2.709	3.095
0.500	0.784	0.794	2.766	3.175
0.600	0.764	0.775	3.043	3.567
0.700	0.746	0.760	3.308	3.951
0.800	0.730	0.746	3.562	4.327
0.900	0.716	0.734	3.807	4.697
1.000	0.703	0.723	4.044	5.061
1.500	0.653	0.681	5.141	6.813
2.000	0.617	0.652	6.130	8.478
2.500	0.589	0.630	7.045	10.079
3.000	0.567	0.612	7.905	11.631

of the Birch equation of state. Thus equation 8 may be used to extrapolate to moderately high pressure. Equation 18 should be used to extrapolate to high pressure.

TEMPERATURE CORRECTION

Comparison of the seismically observed values of ϕ_{FLD} for the earth with laboratory data must be done at the same reference temperature. Consider a material suspected to occur at a depth where the temperature is T and the pressure P. The values of K_0, V_0, and ρ_0 determined in the laboratory at temperature t (likely room temperature) and zero pressure must be extrapolated to T and P. Either equation 8 or equation 18, as appropriate for the pressure, both of which are expressed here as the adiabatic equations of state, may be used. The generalization of these equations to an arbitrary temperature T follows a formalism presented by *Gilvarry* [1957, 1962] and involves replacing K_0, V_0, and ρ_0 (the laboratory temperature measurements) by $k(T)$, $v(T)$, and $\rho(T)$ defined as

$$k(T) = K_0 \exp\left[-\int_t^T \psi_0 \alpha_0 \, dT\right]$$

$$v(T) = V_0 \exp\left[\int_t^T \alpha_0 \, dT\right] \tag{19}$$

$$\rho(T) = \rho_0 \exp\left[-\int_t^T \alpha_0 \, dT\right]$$

where α_0 is the coefficient of volume expansion evaluated at $P = 0$ and ψ_0 is an anharmonic parameter arising from temperature effects given by

$$\psi_0 = \psi_T = -\frac{1}{\alpha}\left(\frac{\partial \ln K}{\partial T}\right)_P \tag{20}$$

At high temperature, where the specific heat at constant volume approaches the Dulong-Petit limit,

$$\psi_T = \psi_s + \gamma_G \tag{21}$$

where γ_G is Grüneisen's ratio and

$$\psi_s = -1/\alpha(\partial \ln K_s/\partial T)_P \tag{22}$$

as given originally by *Grüneisen* [1912, p. 278].

With this temperature correction, the tables in the appendix are still applicable if we understand the pressures to be $P/k(T)$, the relative volumes to be $[V/v(T)]_s$, and the normalized seismic parameter to be $[\phi(P, T)/\phi(T)]_s$ where $\phi(T) \equiv k(T)/\rho(T)$.

APPENDIX

Comparison of volume and the seismic parameter ϕ as a function of pressure for different values of m. For most solids, value of m ranges from 4 to 6; Tables 1, 2, and 3 below are useful for estimating the seismic parameter (as well as volume) whenever K_0 and m are known for a solid under discussion.

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