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Comperative Justification of Different Equations of State Using Pressure Volume Relationship



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Abstract

The behavior of solids has been studied in many ways and after these studies we have come to know about various conclusions in terms of the properties of solids. Pressure and Volume relationships play an important role in these studies. Any equation of state is the best tool to study the effect of pressure on the volume of any solid. A number of equations of state are developed for various studies of material. Some theoretical attempts have been made in the present paper to understand that which Equation of State is in good agreement with the Experimental results.

Keywords: EOS, Pressure, Volume.

Introduction

High pressure research is a field of enormous relevance both for its scientific interest and for its industrial and technological applications. Many materials undergo fascinating changes in their physical and chemical characteristics when subjected to extreme pressure. This behavior is caused by the involvement in bonding of electrons that would otherwise not be chemically active under zero-pressure conditions. By and large, current chemical knowledge and the traditional rules for valence electrons are at a loss to explain most of the changes induced when materials are compressed, which makes (chemical bonding under pressure an exciting research topic that has received much attention in recent years).

The study of pressure effects can be approached in two mostly complementary ways: experimentally and computationally. In both cases, the basic object under study is the change in crystal structure a material undergoes when a given pressure and temperature are applied as shown in its thermodynamic pressure-temperature phase diagram. Experimentally, the application of temperature is relatively straightforward, but imposing high pressure on a sample requires specialized techniques that have been under development for the past 80 years. The field of experimental high-pressure physics was pioneered by Percy Bridgman, who received the Nobel Prize for his efforts in 1946.

Objectives of the Study

High pressure is a very active field of research and one whose ramifications affect many scientific and technological fields, from astrophysics and geophysics to materials physics and the food industry. From the fundamental view, the behavior of materials at high pressure is still poorly understood, and the usual textbook chemistry rules that apply to zero-pressure chemistry are essentially useless when molecules and materials are subjected.. Calculated data (elastic moduli, longitudinal and shear velocities, thermal expansion and conduction, heat capacities, Grüneisen parameters, transition pressures, Clapeyron slopes, etc.) are very helpful in the interpretation of experimental data and in the construction of models of the Earth's interior. In order to describe the thermodynamic state of a system, certain variables are required.

Review of Literature

The thermodynamic state of a solid can be specified by its pressure (P), volume (V) and temperature (T). Out of these three variables P, V and T only two are independent variables and the third may be considered as the function of the remaining two. The relationship between the pressure P, the volume V and absolute temperature T for the substance is known as its equation of state (EOS). Thermodynamic properties of solids under high pressure and high temperature can be understood by investigating an adequate form of EOS. An equation of state

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can be used to determine the bulk modulus K and its variation with pressure and temperature [1,2,8]. These equations can in principle be derived from the knowledge of interatomic potentials for solids. Phenomenological forms of equation of state can also be derived by considering some particular relationship for the pressure dependence and temperature dependence of bulk modulus. The isothermal bulk modulus (K) can be determined from an isothermal equation of state representing relation between pressure and volume at fixed temperature. There exist a number of such equations obtained by various investigations [3]. An isobaric equation of state yields the relation between volume and temperature at constant pressure. The Murnaghan's EOS and the Birch-Murnaghan EOS based on the finite strain theories have been widely used in the field of high pressure physics of solids. The relationship between the pressure and the corresponding change in volume provides an important information regarding the compression of solids under the effect of pressure. The Murnaghan's equation as well as Birch-Murnaghan equation need to be modified by incorporating the higher order terms if we have to predict pressure-volume relationship at simultaneously elevated temperatures and pressures. The Birch-Murnaghan equation of state is based on the finite strain theory using the Eulerian strain whereas the Murnaghan's equation of state is based on the assumption for the linear dependence of bulk modulus on pressure.

Concept and Hypothesis

In this study we have presented the formulations used and numerical analysis for various solids. The results obtained in present study are discussed and compared with available experimental data. We study pressure-volume relationship using different equations of state for NaCl and MgO. This provides an opportunity for testing various equation of state corresponding to a wide range of compressions. In the present study, we consider eight equations of state(1-8). Reliable values of compressions (V/V₀) have been obtained by Birch for NaCl at different pressures by making a careful analysis of experimental measurements. This provides an opportunity for testing various equation of state corresponding to a wide range of compressions.

These equations of state yield following relationships between pressure P and compression

Tait's Equation of state

$$P = \frac{K_0}{K_0^1} \left[\left\{ \exp(K_0^1 + 1) \left(1 - \frac{V}{V_0} \right) \right\} - 1 \right] \dots\dots(1)$$

Born – Mie Equation of state

$$P = \frac{3K_0}{3K_0^1 - 8} \left(\left(\frac{V}{V_0} \right)^{4/3 - K_0^1} - \left(\frac{V}{V_0} \right)^{-4/3} \right) \dots\dots(2)$$

Brennan – Stacey Equation of state

$$P = \frac{3K_0 (V/V_0)^{-4/3}}{(3K_0^1 - 5)} \left[\left\{ \exp \left(\frac{3K_0^1 - 5}{3} \right) \left(1 - \frac{V}{V_0} \right) \right\} - 1 \right] \dots\dots(3)$$

Shanker *et al* Equation of state

$$P = \frac{3K_0 (V/V_0)^{-4/3}}{(3K_0^1 - 8)} \left\{ \left(1 - \frac{1}{t} + \frac{2}{t^2} \right) \right\} \{ \exp.(ty) - 1 \} + y \left\{ 1 + y - \frac{2}{t} \right\} \exp(ty) \dots\dots(4)$$

Where $y = 1 - \frac{V}{V_0}$ and $t = K_0^1 - 8/3$

Vinet *et al* Equation of state

$$P = \frac{3K_0 (1 - V/V_0)^{-1/3}}{(V/V_0)^{2/3}} \exp \left[\frac{3}{2} (K_0^1 - 1) \left(1 - \frac{V}{V_0} \right) \right] \dots\dots(5)$$

Birch – Murnaghan equation of state

$$P = \frac{3K_0}{2} \left(\left(\frac{V}{V_0} \right)^{-7/3} - \left(\frac{V}{V_0} \right)^{-5/3} \right) \dots\dots(6)$$

Born-Mayer equation of state

$$P = \frac{3K_0}{\sigma - 2} \left[\left(\frac{V}{V_0} \right)^{-2/3} \exp \left\{ \sigma \left(1 - \left(\frac{V}{V_0} \right)^{-1/3} - \left(\frac{V}{V_0} \right)^{-4/3} \right) \right\} \right] \dots\dots(7)$$

Murnaghan's equation of state

$$P = \frac{K_0}{K_0^1} \left\{ \left(\frac{V}{V_0} \right)^{-K_0^1} - 1 \right\} \dots\dots(8)$$

As we know that the compressibility of any material gives the information about its crystal structure we will have to develop a relation between bulk modulus and pressure derivatives. Expressions for isothermal bulk modulus are obtained using the relationship $K_T = -V (dP/dV)_T$. The corresponding expressions are

Tait's equation of state

$$K = K_0 \frac{V}{V_0} \left[\left\{ \exp(K_0^1 + 1) \left(1 - \frac{V}{V_0} \right) \right\} \right] \dots\dots(9)$$

Born – Mie equation of state

$$K = K_0 \left(\left(\frac{V}{V_0} \right)^{4/3 - K_0^1} + \frac{4}{3} P \right) \dots\dots(10)$$

Brennan – Stacey equation of state

$$K = K_0 \left(\frac{V}{V_0} \right)^{1/3} \left[\left\{ \exp \left(K_0^1 - \frac{5}{3} \right) \left(1 - \frac{V}{V_0} \right) \right\} + \frac{4}{3} P \right] \dots\dots(11)$$

Shanker *et al* Equation of state

$$K = K_0 \left(\frac{V}{V_0} \right)^{1/3} \exp \left\{ \left(K_0^1 - \frac{5}{3} \right) \left(1 - \frac{V}{V_0} \right) \right\} + \frac{4}{3} P \dots\dots(12)$$

Vinet *et al* equation of state

$$K = Kx^2 [1 + (\eta x + 1)(1 - x)] \exp\{\eta(1 - x)\} \dots\dots(13)$$

Where $x = \left(\frac{V}{V_0} \right)^{1/3}$ and $\eta = \frac{3}{2} (K_0^1 - 1)$

Birch – Murnaghan equation of state

$$K = \frac{1}{2} K_0 (7X^{-7} - 5X^{-5}) + \frac{3}{8} K_0 (K_0^1 - 4) (9X^{-9} - 14X^{-7} + 5X^{-5}) \dots\dots(14)$$

Murnaghan equation of state

$$K = K_0 \left(\frac{V}{V_0} \right)^{-K_0^1} \dots\dots(15)$$

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Research Design

The different equations (1) to (15) contains only two parameters K_0 and K_0^1 both at zero pressure. It has been the usual practice to adjust the parameters K_0 and K_0^1 in order to achieve the agreement with experimental data. This procedure of fitting does not provide any useful insight for the physics of equations of state. In order to make a real test of various equations of state, we have used experimental values $K_0 = 23.84$ G Pa and $K_0^1 = 5.35$ for NaCl measured ultrasonically. For presenting a meaningful comparison of results obtained from various equations, we have used the same value of input parameters without making any adjustments.

A comparison of results obtained from different equations of state has been presented in Table 1. The comparison reveals that Murnaghan's equation of state (equation 8) yields largest deviations from experimental data. This demonstrate that the assumption for the linear dependence of isothermal

bulk modulus on pressure is inadequate, and therefore one should consider the higher order terms showing the non-linear dependence of bulk modulus on pressure. The second largest deviations are found in case of the Born-Mie equation (2). The Birch-Murnaghan's equation (6) and the Born-Mayer equation (7) yields the results which are close to each other, the deviations at the maximum compression are about 15 percent for both equation (6) and (7). In low pressure region (upto 5 GPa) all the equations yield almost similar results. The deviations go on increasing with the increase in compression ($1 - V/V_0$). At the highest compression, it is found that the usual Tait's equation (1), the Brennen- Stacey equation (3), the Vinet *et al* equation (5) and Shanker *et al* equation (4) yield results which are in agreement with the experimental data within 10 percent. The Brennen-Stacey equation (3) gives the best agreement with P-V data based on experimental measurements.

Table-1 Values of Pressure (GPa) as a function of V/V_0 calculated from different EOS for NaCl

| V/V0 | EXP. Value | Taits eq | Born mie eq. | Bernnan eq | Shanker et al | Vinet et al eq | Birch murnaghan eq | Born mayer eq | Murnaghan eq |
|-------|------------|----------|--------------|------------|---------------|----------------|--------------------|---------------|--------------|
| .9627 | 1 | 1.00 | 1.01 | 1.00 | 1.00 | 1.00 | 1.00 | 1.00 | 1.01 |
| .9324 | 2 | .02 | 2.02 | 2.01 | 2.01 | 2.01 | 2.01 | 2.01 | 2.02 |
| .9067 | 3 | 3.07 | 3.04 | 3.03 | 3.03 | 3.03 | 3.04 | 3.04 | 3.07 |
| .8845 | 4 | 4.14 | 4.08 | 4.04 | 4.05 | 4.04 | 4.06 | 4.07 | 4.14 |
| .8649 | 5 | 5.23 | 5.13 | 5.06 | 5.08 | 5.06 | 5.10 | 5.11 | 5.23 |
| .7910 | 10 | 11.2 | 10.6 | 10.3 | 10.3 | 10.3 | 10.5 | 10.5 | 11.2 |
| .7397 | 15 | 17.9 | 16.6 | 15.6 | 15.7 | 15.6 | 16.1 | 16.2 | 17.9 |
| .7004 | 20 | 25.5 | 22.9 | 21 | 21.2 | 21.1 | 22.1 | 22.1 | 25.5 |
| .6685 | 25 | 34.0 | 29.6 | 26.7 | 26.9 | 26.7 | 28.3 | 28.4 | 34.0 |
| .6416 | 30 | 43.4 | 36.8 | 32.1 | 32.7 | 32.5 | 34.8 | 34.9 | 43.4 |

The Brennen-Stacey equation (3) gives the best agreement with P-V data based on experimental measurements which is based on the free volume formula for the pressure dependence of the Gruneisen parameter γ , which can be expressed as follows

$$\gamma = - \frac{(1/2)(dK/dP)-(5/6)+(2/9)P/B}{1-(4/3)P/B} \dots\dots\dots(16)$$

The volume dependence of Gruneisen parameter given by empirical relationship

$$\gamma = \gamma_0 (V/V_0)^q \dots\dots\dots(17)$$

Where, q is known as the second Gruneisen parameter and is also expressed as

$$q = \frac{V}{\gamma} \left(\frac{d\gamma}{dV} \right) \dots\dots\dots(18)$$

Using equations (16) and (18), and taking the Gruneisen parameter γ to be directly proportional to volume V, i.e., $q = 1$, Brennen and Stacey have derived their equation of state given by equation (3). The condition $q = 1$ is well satisfied in case of NaCl crystal. This explains why the Brennan-Stacey equation of state yields the best agreement with the experimental compression data. Equation (16) for γ is reduced to the following form at $P = 0$

$$\gamma_0 = \frac{1}{2} \left(K_0^1 - \frac{5}{3} \right) \dots\dots\dots(19)$$

Equation (19) yields $\gamma_0 = 1.84$ when $K_0^1 = 5.35$. This value is in good agreement with the

experimental data [4.5]. It is also pertinent to mention here that $q = 1$ leads to the following results

$$\delta_T = \frac{dK}{dP} \dots\dots\dots(20)$$

$$\left(\frac{\delta K}{\delta T} \right)_V = 0 \dots\dots\dots(21)$$

And

$$\gamma/V = \text{constant} \dots\dots\dots(22)$$

Where, δ_T is the Anderson-Gruneisen parameter. The experimental thermoelstic data available for NaCl crystal [6] satisfy the relationships (20), (21) and (22) almost exactly. Thus the basic assumptions, on which the Brennan-Stacey equation (3) is based, are well supported by experimental data.

Thermoelastic properties of MgO at high temperatures and high pressures using the potential induced breathing (PIB) electron gas model based on first principles approach has been discussed earlier[7,9-12]. The ab-initio method of Isaak *et al* based on the detailed calculations of the Helmholtz energy F versus volume V at constant temperature T at selected temperatures. By taking appropriate derivatives of F, values of pressure P, isothermal bulk modulus K_T and its pressure derivatives K_T^1 are numerically determined.

We use the ab-initio values at $P = 0$ obtained by Isaak *et al* for MgO at 300 K, 500 K, 1000 K, 1500 K and 2000 K as reported by Anderson given in Table 2. and used as input for all the equations of state

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under study without making any adjustment. The results for pressure P, isothermal bulk modulus K_T

obtained for different equations are compared with ab-initio values in Table (3) and (4).

Table-2 : Values of Input Data on $K_0 = K_T$ and K_T^1 for MgO Based on the PIB Model

| Temperature (K) | K_0 (GPa) | K_0^1 (dimensionless) |
|-----------------|-------------|-------------------------|
| 300 | 180 | 4.15 |
| 500 | 175 | 4.21 |
| 1000 | 160 | 4.36 |
| 1500 | 144 | 4.53 |
| 2000 | 128 | 4.74 |

Table 3: Values of pressure P(GPa) for MgO at different compressions

| V/V_0 T=300K | (a) | (b) | © | (d) | (e) | (f) | (g) | V/V_0 T=500K | (a) | (b) | © | (d) | (e) | (f) | (g) |
|-------------------|------|------|------|------|------|------|------|-------------------|------|------|------|------|------|------|-----|
| | 1.0 | 0 | 0 | 0 | 0 | 0 | 0 | | 0 | 1.0 | 0 | 0 | 0 | 0 | 0 |
| .95 | 10.3 | 10.3 | 10.3 | 10.3 | 10.3 | 10.3 | 10.3 | .95 | 10 | 10 | 9.99 | 10 | 10 | 10 | 10 |
| .90 | 23.5 | 23.6 | 23.5 | 23.6 | 23.5 | 23.6 | 24.0 | .90 | 23 | 23.1 | 22.9 | 23 | 23 | 23 | 23 |
| .85 | 40.7 | 41.1 | 40.6 | 40.8 | 40.7 | 41.7 | 41.0 | .85 | 39.8 | 40.2 | 39.7 | 39.8 | 39.8 | 40.1 | 40 |
| .80 | 62.9 | 64.1 | 62.8 | 63.2 | 63.0 | 64.0 | 64.0 | .80 | 61.6 | 62.8 | 61.4 | 61.8 | 61.7 | 62.6 | 62 |
| .75 | 91.7 | 94.8 | 91.5 | 92.4 | 92.1 | 94.4 | 94.0 | .75 | 90 | 93 | 89.7 | 90.6 | 90.3 | 92.6 | 91 |
| .70 | 129 | 136 | 129 | 131 | 130 | 135 | 133 | .70 | 127 | 134 | 127 | 128 | 128 | 133 | 130 |
| .65 | 177 | 193 | 178 | 182 | 181 | 191 | 187 | .65 | 175 | 190 | 175 | 179 | 178 | 188 | 183 |
| .60 | 239 | 272 | 244 | 249 | 249 | 268 | 260 | .60 | 236 | 269 | 240 | 245 | 246 | 265 | 255 |

| V/V_0 T=100 0K | (a) | (b) | © | (d) | (e) | (f) | (g) | V/V_0 T=1500 K | (a) | (b) | © | (d) | (e) | (f) | (g) |
|------------------------|------|------|------|------|------|------|-----|------------------------|------|------|------|------|------|------|-----|
| | 1.0 | 0 | 0 | 0 | 0 | 0 | 0 | | 0 | 1.0 | 0 | 0 | 0 | 0 | 0 |
| .95 | 9.17 | 9.8 | 9.17 | 9.17 | 9.17 | 9.18 | 9.2 | .95 | 8.29 | 8.30 | 8.29 | 8.29 | 8.29 | 8.30 | 8.3 |
| .90 | 21.2 | 21.2 | 21.1 | 21.2 | 21.1 | 21.1 | 21 | .90 | 19.2 | 19.3 | 19.2 | 19.2 | 19.2 | 19.3 | 19 |
| .85 | 36.9 | 37.2 | 36.7 | 36.9 | 36.8 | 37.1 | 37 | .85 | 33.7 | 33.9 | 33.5 | 33.6 | 33.6 | 33.9 | 34 |
| .80 | 57.4 | 58.4 | 57.1 | 57.4 | 57.3 | 58.3 | 58 | .80 | 52.7 | 53.7 | 52.3 | 52.7 | 52.5 | 53.5 | 53 |
| .75 | 84.2 | 87 | 83.8 | 84.6 | 84.3 | 86.6 | 85 | .75 | 77.7 | 80.4 | 77.2 | 77.9 | 77.7 | 80 | 79 |
| .70 | 119 | 126 | 119 | 120 | 120 | 125 | 122 | .70 | 111 | 117 | 110 | 112 | 111 | 116 | 113 |
| .65 | 165 | 180 | 165 | 168 | 168 | 178 | 172 | .65 | 154 | 169 | 154 | 157 | 156 | 167 | 160 |
| .60 | 225 | 257 | 227 | 232 | 233 | 253 | 240 | .60 | 212 | 243 | 213 | 218 | 218 | 238 | 224 |

Table (4) Values of isothermal bulk modulus K_T (GPa) for MgO at different compressions

| $V=V_0$ T=300 0K | (a) | (b) | © | (d) | (e) | (f) | (g) | $T=500$ 00K | (a) | (b) | © | (d) | (e) | (f) | (g) |
|------------------------|-----|------|-----|-----|-----|------|------|----------------|-----|------|-----|-----|-----|------|------|
| | 1.0 | 180 | 180 | 180 | 180 | 180 | 180 | | 180 | 1.0 | 175 | 175 | 175 | 175 | 175 |
| .95 | 221 | 222 | 221 | 221 | 221 | 222 | 222 | .95 | 216 | 216 | 216 | 216 | 216 | 216 | 215 |
| .90 | 271 | 274 | 270 | 272 | 271 | 273 | 273 | .90 | 265 | 268 | 264 | 266 | 265 | 267 | 266 |
| .85 | 331 | 339 | 330 | 334 | 332 | 338 | 336 | .85 | 325 | 333 | 324 | 327 | 325 | 332 | 328 |
| .80 | 403 | 423 | 402 | 410 | 406 | 420 | 415 | .80 | 397 | 416 | 395 | 403 | 399 | 414 | 406 |
| .75 | 489 | 531 | 491 | 506 | 499 | 526 | 515 | .75 | 483 | 524 | 483 | 499 | 419 | 520 | 505 |
| .70 | 591 | 673 | 599 | 626 | 614 | 664 | 643 | .70 | 585 | 667 | 592 | 619 | 606 | 658 | 613 |
| .65 | 710 | 863 | 733 | 779 | 759 | 847 | 806 | .65 | 705 | 858 | 726 | 771 | 751 | 842 | 792 |
| .60 | 847 | 1121 | 901 | 976 | 944 | 1094 | 1019 | .60 | 844 | 1119 | 894 | 968 | 937 | 1092 | 1002 |

| V/V_0 T=100 0 K | (a) | (b) | © | (d) | (e) | (f) | (g) | V/V_0 T=1500K | (a) | (b) | © | (d) | (e) | (f) | (g) |
|-------------------------|-----|------|-----|-----|-----|------|-----|--------------------|-----|------|-----|-----|-----|------|-----|
| | 1.0 | 160 | 160 | 160 | 160 | 160 | 160 | | 160 | 1.0 | 144 | 144 | 144 | 144 | 144 |
| .95 | 199 | 199 | 199 | 199 | 199 | 199 | 199 | .95 | 180 | 181 | 180 | 180 | 180 | 180 | 181 |
| .90 | 246 | 248 | 245 | 246 | 246 | 248 | 246 | .90 | 225 | 227 | 224 | 225 | 225 | 227 | 226 |
| .85 | 304 | 311 | 302 | 305 | 303 | 310 | 306 | .85 | 281 | 287 | 278 | 281 | 280 | 286 | 283 |
| .80 | 374 | 392 | 372 | 379 | 375 | 390 | 381 | .80 | 348 | 366 | 345 | 352 | 348 | 362 | 354 |
| .75 | 458 | 498 | 457 | 471 | 464 | 493 | 476 | .75 | 430 | 468 | 427 | 441 | 434 | 462 | 444 |
| .70 | 559 | 639 | 563 | 588 | 576 | 630 | 597 | .70 | 530 | 607 | 530 | 553 | 542 | 595 | 560 |
| .65 | 679 | 830 | 695 | 738 | 719 | 813 | 753 | .65 | 648 | 796 | 658 | 700 | 681 | 775 | 709 |
| .60 | 819 | 1093 | 860 | 932 | 902 | 1064 | 954 | .60 | 789 | 1061 | 821 | 890 | 861 | 1025 | 901 |

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The comparison reveals that the Vinet equation of state and the Shanker equation of state yield very similar results, which are also close to the ab-initio values. On other hand, the results based on the remaining equation of state deviate substantially from the ab-initio values for P and K_T . It is pertinent to mention here that the values determined from the PIB model have been found to present good agreement with experimental data on thermoelastic properties at high pressures and at high temperatures. Thus the Vinet equation of state and the Shanker equation of state, which have been found to yield good agreement with PIB model. [Table (3)] are also consistent with the thermoelastic properties. The present study supports the view point that phenomenological isothermal equation of state of solids can be applicable even at high temperatures if the parameters V_0 , K_0 and so forth, estimated at that temperature are used.

Conclusion

The comparison reveals that the Vinet equation of state and the Shanker equation of state yield very similar results, which are also close to the ab-initio values. On other hand, the results based on the remaining equation of state deviate substantially from the ab-initio values for P and K_T . It is pertinent to mention here that the values determined from the PIB model have been found to present good agreement with experimental data on thermoelastic properties at high pressures and at high temperatures. Thus the Vinet equation of state and the Shanker equation of state, which have been found to yield good agreement with PIB model. are also consistent with the

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