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Assessment of Several Equations of State for the Calculation of Thermodynamic Coefficients of Solids

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This comprehensive study examines five equations of state (EoSs)-Vinet, Kholiya, Murnaghan, Birch-Murnaghan, and the newly derived Srivastava-Pandey EoS. Bulk modulus and thermal expansion coefficient expressions were derived, and compression versus pressure and thermal expansion curves were plotted. The results, analysed across four materials, provide a thorough understanding of the subject. Pressure and bulk modulus increased with compression, while thermal expansion decreased for MgO. Srivastava-Pandey and Vinet EoS emerged as the most accurate, matching experimental data, with other EoS deviating at high compression. NaF exhibited similar trends, with Vinet, Srivastava-Pandey, and Birch-Murnaghan closely aligning with experimental results, though Birch-Murnaghan showed deviations at high compression. In Xe, Kholiya, and Birch-Murnaghan EoS were better suited, producing thermal expansion results that followed the experimental pattern. The Srivastava-Pandey EoS produced the most accurate results for Cu, while the Murnaghan model showed significant deviations in thermal expansion. The Shanker-Kumar criterion validates the thermal expansion coefficient, providing a robust foundation for further research and practical applications.

Keywords: Equation of state, Thermal expansion coefficient, Compression, Bulk modulus

INTRODUCTION

This study holds significant value across various fields, including geophysics, material science, condensed matter physics, and high-pressure physics. It enhances our understanding of material behaviour, particularly in extreme environments where the volume thermal expansion coefficient, a critical property under standard ambient conditions, can vary significantly with pressure and compression. This detailed analysis is essential for various applications, from engineering to planetary science [1-5].

The thermal expansion coefficient, a crucial indicator of how a material responds to changes in high pressure and temperature, is not just theoretical. It provides practical insight into how much a material's volume changes with temperature variations, thereby helping to assess its structural integrity and stability, especially under diverse and extreme environmental conditions. Understanding these factors is crucial for predicting and managing material performance [6,7,8].

Different assumptions are suitable under various pressures and temperatures. Models such as Murnaghan, Birch-Murnaghan, and Vinet equations of state (EoS) each have specific applications. The Murnaghan EoS is ideal for moderate pressures. At the same time, the Birch-Murnaghan and Vinet EoS are better suited for higher pressures, with the Vinet EoS offering a broader range of applicability. Insights from these models play a crucial role in improving the accuracy of compression-dependent expansion coefficients

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under extreme conditions [9-12].

This study employs several EoS models, including the Vinet, Birch-Murnaghan, Kholiya, Murnaghan, and Srivastava-Pande models, to determine which one best predicts compression-dependent expansion coefficients at high pressures [13-15]. The analysis sheds light on how compression impacts materials, which supports material design for extreme conditions and advances predictive modelling in high-pressure physics and planetary sciences. These findings enhance our theoretical understanding and inspire practical applications, ranging from designing materials for space exploration to developing engineering solutions for high-pressure environments.

The study calculates these coefficients by examining copper (Cu), magnesium oxide (MgO), xenon (Xe), and sodium fluoride (NaF), covering a range of material types: copper as a metal, magnesium oxide as a semiconductor, xenon as a noble gas, and sodium fluoride as an ionic halide.

COMPUTATIONAL METHODOLOGY

Methodology

The Vinet EoS, often called the universal EoS, is derived based on the solids' interatomic potential and compressibility. It works at the low and middle-pressure ranges, but deviation occurs at high compression. Vinet EoS is expressed as [16]:

$$P = 3K_0 x^{-2}\left(1-x\right)\exp\left\{\varepsilon\left(1-x\right)\right\} \tag{1}$$

where

$$x = \left(\frac{V}{V_0}\right)^{\frac{1}{3}} \text{ and } \varepsilon = \frac{3}{2}\left(K_0^{'}-1\right)$$

The Kholiya Equation of State (EoS) is based on a virial theorem. While initially derived for nanomaterials, it is also applicable to other solids [17].

$$P = \frac{K_0}{2}\left[\left(K_0^{'}-3\right)-2\left(K_0^{'}-2\right)\left(x\right)^{-3}+\left(K_0^{'}-1\right)\left(x\right)^{-6}\right] \tag{2}$$

Where

$$x = \left(V/V_0\right)^{1/3}$$

The Murnaghan equation of state is the simplest form of the equation of state derived by considering the bulk modulus as a function of pressure. It works well at low compression, but deviation occurs at high compression. Murnaghan EoS can be expressed as [17]:

$$P = \frac{K_0}{K_0^{'}}\left[\left(x^3\right)^{-K_0^{'}}-1\right] \tag{3}$$

Where

$$x = \left(V/V_0\right)^{1/3}$$

The Birch-Murnaghan Equation of State is a widely used equation of state with high accuracy at high compression. It is derived from finite strain theory, and its applicability at low compression yields deviations; however, at high compression, accuracy improves. Birch-Murnaghan EoS can be expressed as [18]:

$$P = \frac{3}{2}K_0[x^{-7}-x^{-5}]\left[1+\frac{3}{4}\left(K_0^{'}-4\right)\left(x^{-2}-1\right)\right] \tag{4}$$

Where

$$x = \left(V/V_0\right)^{1/3}$$

The Srivastava-Pandey Equation of State is the first EoS to consider the third order of compression and anharmonic effects at high compression. It operates with high accuracy at both low and high compression levels. The Srivastava-Pandey EoS can be expressed as [17-18]:

$$P = K_0\left(x\right)^{-4}\left[\frac{\left\{\alpha^3(1+y+y^2+y^3)+\alpha^2(-3y^2-2y-1)+\alpha(6y+2)-6\right\}e^{\alpha y}-(\alpha^3-\alpha^2+2\alpha-6)}{\alpha^4}\right] \tag{5}$$

Where $y = 1-x^3$ and $\alpha = \frac{3K_0^{'}-8}{3}$.

The bulk modulus of solids can be expressed as:

$$K_T = -V\frac{\partial P}{\partial V} \tag{6}$$

Using (7) on (1), (2), (3), (4), (5), and (6), we can find an expression for the bulk modulus at the pressure P for considering the equation of state:

$$K_T = K_0 x^{-2}\left[1+\left\{(1+\eta x)(1-x)\right\}\right]\exp\eta(1-x) \tag{7}$$

$$K_T = K_0\left(K_0^{'}-2\right)\left(\frac{V}{V_0}\right)^{-1}\left[\left(\frac{K_0^{'}-1}{K_0^{'}-2}\right)\left(\frac{V}{V_0}\right)^{-1}-1\right] \tag{8}$$

$$K_T = K_0\left(\frac{V}{V_0}\right)^{-K_0^{'}} \tag{9}$$

$$K_T = \frac{K_0}{2}\left[7x^{-7}-5x^{-5}\right]+\frac{3}{8}K_0\left(K_0^{'}-4\right)\left(9x^{-9}-14x^{-7}+5x^{-5}\right) \tag{10}$$

$$K_T = K_0\left(\frac{V}{V_0}\right)^{-1/3}\left(1+y+y^2+y^3\right)\exp\left(\alpha y\right)+\frac{4}{3}P \tag{11}$$

Volume Thermal Expansion Coefficient

The product of the volumetric thermal expansion coefficient (α) and isothermal bulk modulus (K_T) is a proven and unchanging constant for all solids. This scientific principle has been extensively studied and verified by experts in the field. By knowing and confidently applying this principle, we can gain deeper insights into the behaviour of materials under varying temperature and pressure conditions and open new pathways to advances in material science.

$$\alpha K_T = \text{Constant}$$

Shanker and Kumar show that [19]:

$$\alpha K_T = \alpha_0 K_0$$

$$\alpha = \alpha_0\left(\frac{K_0}{K_T}\right) \tag{12}$$

Lattice softening tends to occur as the temperature rises, and this usually leads to a decrease in the bulk modulus. The thermal expansion coefficient, conversely, goes up. Equation (12) captures this inverse relationship, linking these properties. This relationship is beneficial for verifying the thermal expansion data obtained from first-principles calculations and the equation of state. It is a validation tool. If your calculated α(T) and K_T don't line up with Eq. (12), that could be a signal. Maybe there is something off in your theoretical model. Alternatively, you may need to account for other factors, such as anharmonic effects. Equation (12) can even help in estimating thermal behavior from computed mechanical properties, which is helpful when you don't have experimental thermal expansion data. Ultimately, using this equation helps ensure the thermodynamic coherence of your simulation results, and significantly, it boosts our confidence in how well we predict the behavior of materials at different temperatures. Equation (12) represents the Shanker-Kumar criterion.

Using Eqs. (7), (8), (9), (10), (11), and (12), we calculate the volume thermal expansion coefficient at different compressions.

1. RESULT AND DISCUSSION

In the present study, we have considered five equations of state, of which four (Vinet EoS, Kholiya EoS, Murnaghan EoS, and Birch-Murnaghan EoS) were previously derived, and a new equation of state, Srivastava-Pandey's equation of state, has been recently derived [18]. These equations are represented by Eqs. (1), (2), (3), (4), and (5). After that, we derive an expression for the bulk modulus using Eq. (6), and the expressions corresponding to these equations are represented by (7), (8), (9), (10), and (11). Finally, we calculate the compression-dependent thermal expansion coefficient by using Eq. (12). We draw the curves between compression and pressure, as well as between compression and the thermal expansion coefficient, for better clarity, as shown in Figs. 1-8. The result analysis is divided into four parts, corresponding to each element. The input values used in the calculation are listed in Table 1.

Table 1. Input Parameters Used in the Calculation, such as Bulk Modulus, Pressure Derivative of Bulk Modulus, and Volume Thermal Expansion Coefficient at Zero Pressure and Room Temperature

Elements	K_0 (GPa)	K_0'	$\alpha \times 10^{-5}$ K^{-1}
MgO	173.0 [20]	4.15 [20]	104.2 [24]
NaF	46.50 [21,22]	5.28 [21,22]	9.60 [21,22]
Xe	3.65 [23]	8.87 [23]	174.0 [25]
Cu	130.92 [24]	3.88 [24]	167.0 [27]

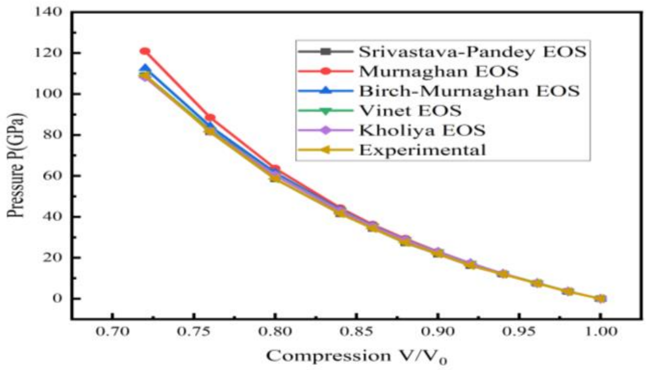

Fig. 1. Variation of pressure concerning compression for MgO with experimental values [24].

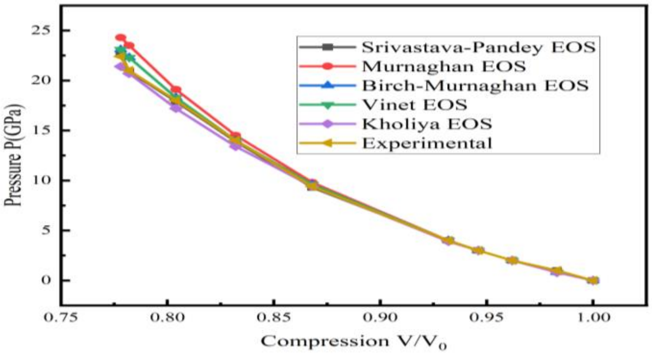

Fig. 2. Variation of pressure concerning compression for NaF with experimental values [28].

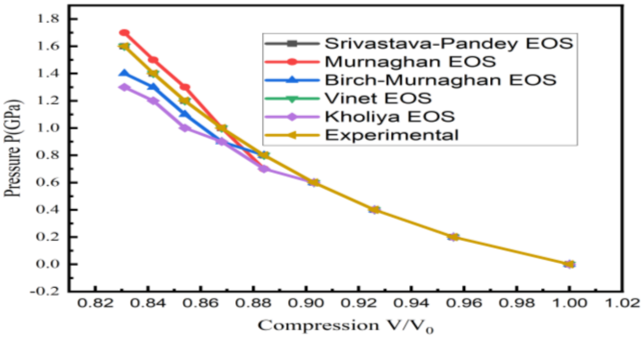

Fig. 3. Variation of pressure with compression for Xe with experimental values [18].

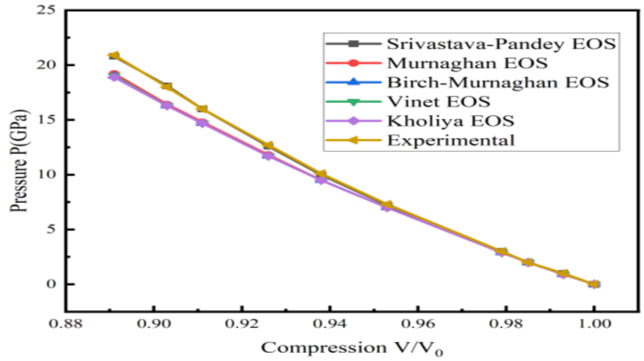

Fig. 4. Variation of pressure concerning compression for Cu with experimental values [28].

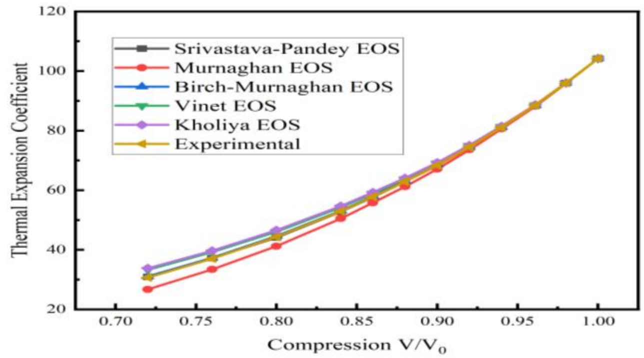

Fig. 5. Variation of thermal expansion coefficient concerning compression for MgO with experimental values [24].

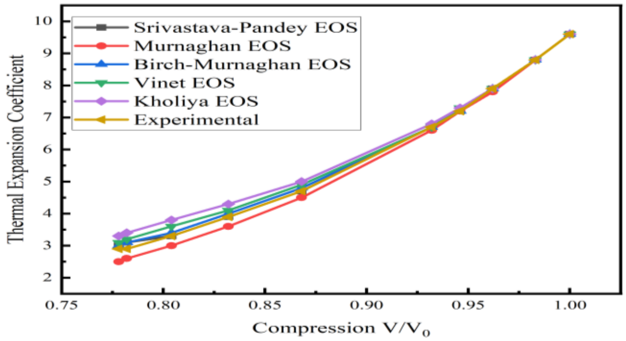

Fig. 6. Variation of thermal expansion coefficient concerning compression for NaF with experimental values [25].

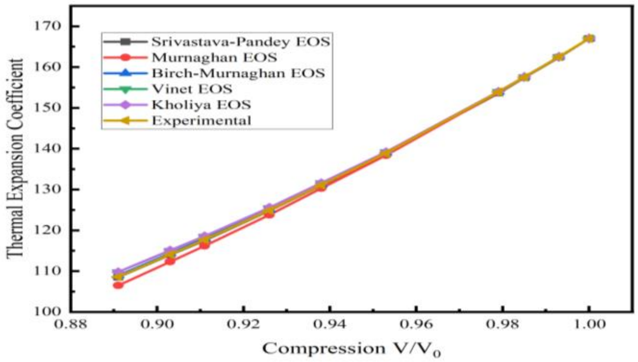

Fig. 7. Variation of thermal expansion coefficient concerning compression for Xe with experimental values [26].

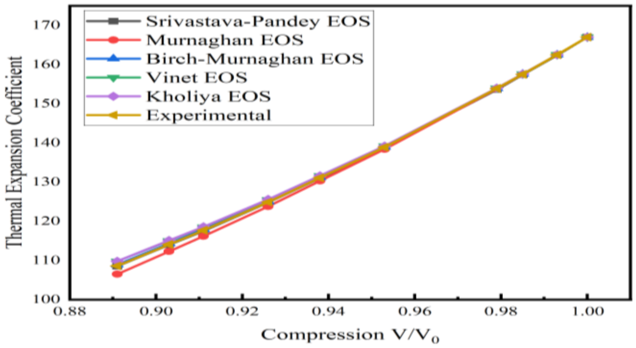

Fig. 8. Variation of thermal expansion coefficient concerning compression for Cu with experimental values [27].

Analysis of MgO

In the case of MgO, the pressure increases, and the bulk modulus increases with increasing compression; however, the thermal expansion coefficient decreases with increasing compression. The calculated pressure values are similar to the experimental data for the Srivastava-Pandey EoS and Vinet EoS, but deviations occur in other state equations at higher compression. The Srivastava-Pandey EoS and Vinet EoS produce results similar to the experimental ones. The equation (Srivastava-Pandey and Vinet EoS) yields the best results for compression-dependent pressure and creates a thermal expansion coefficient that is similar to the experimental data.

Analysis of NaF

In the case of NaF, it is also observed that with increasing compression, pressure and bulk modulus increase. At the same time, the thermal expansion coefficient decreases, following a similar trend to the equation of state. The compression-dependent pressures calculated by the Vinet EoS, Srivastava-Pandey EoS, and Birch-Murnaghan EoS produce results that are identical to the experimental ones; however, the Birch-Murnaghan EoS yields significant deviations from the experimental values. The Kholiya EoS produces results for pressure, which decrease with increasing pressure, whereas the Murnaghan EoS increases with an increase in compression, as observed in experimental data. Regarding the thermal expansion coefficient, the Vinet EoS and Srivastava-Pandey EoS produced results that were similar to the experimental data, and the Birch-Murnaghan EoS also yielded very close results. The thermal expansion coefficient calculated by the Murnaghan EoS increases from the experimental value, and the result produced by Kholiya decreases with increasing compression.

Analysis of Xe

In the case of NaF, it is also observed that with increasing compression, pressure and bulk modulus increase. At the same time, the thermal expansion coefficient decreases, following a similar trend to the equation of state and fulfilling the Stacey criterion. The curves drawn for Kholiya EoS and Birch-Murnaghan EoS show behaviors that differ from those of others. That is, Birch-Murnaghan and Kholiya EoS are fit for rare gas solids. The thermal expansion coefficients calculated by the Vinet EoS, Srivastava-Pandey EoS, Birch-Murnaghan EoS, and Kholiya EoS are in good agreement with the experimental values. Still, the Murnaghan EoS deviates from the experimental results.

Analysis of Cu

In the case of Cu, the result obtained by the Srivastava-Pandey EoS is similar to the experimental value, but other EoSs deviate from the experimental value. Cu belongs to the metallic group and has a binding energy and stability greater than other elements. However, in the case of the volume thermal expansion coefficient, the Murnaghan EoS deviates more than the others. Still, the accuracy of Srivastava-Pandey EoS is greater than that of other state equations.

It is seen that the values of the compression-dependent thermal expansion coefficient depend upon the slope of α and K_T, *i.e.* $\frac{dK_T}{d\alpha}$. The slope of the Srivastava-Pandey EoS is greater than that of the other equations of state; therefore, it gives better results than the others. In the case of MgO, NaF, Xe, and Cu, the slope of the Srivastava-Pandey curve is higher than the others; Vinet EoS gives better results with a slightly lower slope of α and K_T. The slope of α and K_T is measured from data calculated in Tables 1-4. Hence, we conclude that compression-dependent thermal expansivity depends on the slope of α and K_T curve.

It is observed that with an increasing order of compression, the accuracy of compression-dependent thermophysical properties also increases. However, as the order of compression continues to rise, the equation of state becomes increasingly complex. Therefore, there is a need for a higher-order compression equation that remains simple. Due to the complexity of the equation of state, the formulation of other properties, such as melting temperature and Debye temperature, deviated from experimental values because terms were neglected to form a quadratic equation, resulting in an inverted type of equation of state.

Discussion of EoS Discrepancies

It is seen that no single Equation of State (EoS) works flawlessly across all materials studied – MgO, NaF, Xe, and Cu. An EoS's accuracy tends to be material-dependent, not a one-size-fits-all situation. Take the Kholiya and Birch-Murnaghan EoSs; they happen to shine with Xe because their equations are better equipped to describe those soft, weakly held solids, ones with noticeable anharmonicity, such as rare gas crystals. Xe has a shallow potential energy surface, making it quite compressible, and the Kholiya and Birch-Murnaghan EoSs deal with those nonlinearities better than the Murnaghan or Vinet EoSs. Still, these two were not pushed heavily in the final summary, mainly because their strength is very much tied to those rare-gas-type systems. When dealing with ionic (NaF) or metallic (Cu) solids, they do not perform as well compared to Srivastava-Pandey and Vinet, which seem to work more broadly. The final thoughts, then, highlighted those models with more reliable accuracy across different materials.

The inaccuracies of the Murnaghan EoS for Cu offer a helpful insight. As a metallic system, Cu, with its abundance of delocalized electrons, behaves differently in terms of pressure and volume than ionic or van der Waals-bonded systems. The Murnaghan EoS, relying too much on a straightforward linear link between bulk modulus and pressure, misses the subtle curve of Cu's P–V relationship, particularly when squeezed tightly. This difference becomes amplified when examining properties derived from how the EoS changes, such as the thermal expansion coefficient, leading to significant and systematic errors. On the other hand, the Srivastava-Pandey and Vinet EoSs, given their more adaptable equation shapes, perform better in considering metallic bonding, and their results are more in line with experimental observations.

CONCLUSION

An examination of compression-dependent thermal expansion was conducted, focusing on magnesium oxide (MgO), sodium fluoride (NaF), xenon (Xe), and copper (Cu). To achieve this, five distinct equations of state-specifically, Vinet, Kholiya, Murnaghan, Birch-Murnaghan, and Srivastava-Pandey were analyzed. For MgO, it was observed that the Srivastava-Pandey and Vinet EoSs demonstrated a strong correlation with available experimental findings. Conversely, the Murnaghan EoS exhibited notable discrepancies, particularly at higher compression rates. Shifting to NaF, the Vinet and Srivastava-Pandey EoSs again delivered precise estimates for both pressure and thermal expansion, while the Birch-Murnaghan performed reasonably well. However, deviations were present in the Murnaghan and Kholiya EoSs. Concerning Xe, a notable alignment with experimental data was apparent across all EoSs, with the singular exception of the Murnaghan model; the Kholiya and Birch-Murnaghan models showed improved performance, given Xe's nature as a rare gas. In the case of Cu, the Srivastava-Pandey EoS offered the most faithful approximations, with other equations (Murnaghan in particular) displaying marked deviations, a reflection of the strong atomic bonds in Cu, which seemingly lend themselves to the Srivastava-Pandey EoS's accurate modelling. In summary, the findings point to a significant impact of the α–K_T curve's slope on the precision of thermal expansion forecasts. A more consistent and steeper slope, such as that found in the Srivastava-Pandey EoS, makes it a more reliable choice for estimating thermal and mechanical behaviors under compressive force. Its simplicity and degree of accuracy make it a solid contender for such modeling.

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