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Compare Halide Melting Curves using the Equation of State and Lindemann's Law for a Comprehensive Analysis

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Abstract:
The primary objective of this work is to accurately predict the equation of state necessary for calculating the pressure-dependent melting curves of alkali halides. In this study, we assert a distinct model for the melting curve utilizing the Murnaghan, Singh and Kao, Shanker, and Usual-Tait equations of state. Our developed model, rigorously tested and compared with Lindemann's model, defines the relationship among pressure, bulk modulus, pressure derivative of bulk modulus, and volume compression. Our findings unequivocally demonstrate that a substantial increase in melting temperature is directly attributed to a significant increment in bulk modulus and a gradual decrement rate in the first-order pressure derivative of bulk modulus. This study provides unparalleled insights into the fundamental understanding of the effect of pressure on melting temperature. The model we have obtained is unequivocally reliable for extrapolating melting temperature to high pressure, giving you confidence in its application.

Keywords: Thermodynamic model, bulk modulus, First-order pressure derivative of bulk modulus, Melting temperature, Equation of state.

1. Introduction:

Our work significantly contributes to the high-pressure melting of solids, both experimentally and theoretically, which has been a significant and pivotal area of research in shock wave physics, geophysics, astrophysics, and nuclear physics. By providing a comprehensive and accurate prediction of the equation of state necessary for calculating the pressure-dependent melting curves of alkali halides, we are advancing the understanding of these fundamental principles in our respective fields, thereby underlining the importance of our research in these crucial areas of physics [1-2]. Over the past two decades, researchers have dedicated their efforts and passion to understanding solids' physical and chemical properties at high pressure and temperature. They have developed various thermodynamic models and utilized advanced mathematical tools like fractional calculus to understand these complex phenomena better, demonstrating the scientific community's unwavering commitment and enthusiasm in this field, which is a testament to the dedication and passion of researchers in our community [3-5]

The equation of state, a fundamental tool in physics, plays a paramount role in our study of high-pressure solids. It provides a framework for understanding how the volume or density of material changes with pressure and temperature. In our research, the equation of state is crucial in determining thermodynamic properties such as volume compression, bulk modulus, and the first-order pressure derivative of bulk modulus, thereby underlining its importance and relevance to our study [6-8]. The equation of state is particularly valuable in our research as it provides values of required parameters in

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ranges where experimental tools cannot operate due to their constraints. Researchers can elucidate the behavior of solids at high pressure using a combination of experimental and theoretical methods. Experimental methodologies such as the diamond-anvil cell method (DAC), X-ray diffraction methods, and shock wave methods are examples. In contrast, theoretical methods used by researchers include molecular dynamics (MDs), simulation methods, ab initio methods, and models based on the Lindemann-Gilvarry law [9-13].

Correctly predicting high-pressure melting curves using theoretical models is a formidable challenge. This is due to the intricate requirements of particle distribution in both liquid and solid phases and the complex details of interatomic forces. Our research underscores these challenges and the intellectual rigor required to overcome them, highlighting the complexity and dedication involved in our study, which is a testament to our research team's intellectual rigor and commitment [14-17].

Our current objective is to derive the most accurate isothermal equation of state by applying various thermodynamic models to establish a suitable formula for calculating the melting temperature. Determining the melting temperature involves input parameters such as the bulk modulus at zero pressure (K_0) and the first-order derivative of the bulk modulus at zero pressure (K_0'). The literature contains the experimental values of these input parameters. The melting temperature formulation can determine the melting temperature of solids across various pressure ranges of alkali halides. Obtained results from different models derived by different equations of state compared with the available theoretical result in the literature.

2. Methodology and Analysis: This section will explain the methodology used in our research, including the theoretical models and equations of state employed and the analysis of the results obtained.

The melting temperature of solids at different ranges of pressure can be obtained by the equation of state (EOS) such as:

2.1. ***Murnaghan Equation of State:*** Murnaghan proposed the equation of state based on the linear dependence of the isothermal bulk modulus on pressure for solids, which may be written as [18]:

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

Where P is compression-dependent pressure, K_0Is the bulk modulus at zero pressure, K_0' is the first-order derivative of bulk modulus at zero pressure and V/V_0 is volume compression.

The bulk modulus is defined as

$$K = -V\left(\frac{dP}{dV}\right)_T \tag{2}$$

Differentiating equation (1) concerning volume, the isothermal bulk modulus K can be expressed as:

$$K = K_0\left(\frac{V}{V_0}\right)^{-K_0^{'}} \tag{3}$$

The first-order derivative of bulk modulus for pressure is given by

$$K^{'} = \left(\frac{dK}{dP}\right) \tag{4}$$

Using equations (3) and (4), the first-order pressure derivative of the bulk modulus is expressed as:

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$$K' = \frac{K_0 K_0'}{K}\left(\frac{V}{V_0}\right)^{-K_0'} \quad (5)$$

The inverted form of the isothermal equation (1) can be expressed in terms of $\frac{V}{V_0}$ as:

$$\frac{V}{V_0} = \left[1 + P\left(\frac{K_0'}{K_0}\right)\right]^{-\frac{1}{K_0'}} \quad (6)$$

Equation (6) calculates volume compression, which is proportional to the pressure.
The thermal pressure P_{th} is expressed as [19-20]:

$$P_{th} = \int_{T_0}^{T} \alpha K dT \quad (7)$$

Where α is thermal expansivity, T_0 is room temperature, and T is elevated temperature.
Integrating equation (7) then thermal pressure can be expressed as:

$$P_{th} = \alpha K (T - T_0) \quad (8)$$

Thermal pressure at zero pressure is expressed as:

$$P_{th} = \alpha_0 K_0 (T - T_0) \quad (9)$$

From equations (6) and (7), the expression for the melting temperature of solids can be obtained as:

$$T_{mo} - T_0 = \frac{K K_0'}{\alpha_0 K_0 K'} \quad (10)$$

To obtain the melting temperature of a solid at pressure P, terms including K_0' and K_0 in equation (10) must be replaced by K' and K then the equation (10) becomes:

$$T_m(P) = T_0 + (T_{m0} - T_0)\frac{K K_0'}{K_0 K'}\left(\frac{V}{V_0}\right) \quad (11)$$

2.2. ***Singh and Kao Equation of state:*** The Singh and Kao equation of state is expressed as [21]:

$$P = K_0\left(1 - \frac{V}{V_0}\right) + \left\{\frac{K_0(K_0' + 1)}{2}\right\}\left(1 - \frac{V}{V_0}\right)^2 \quad (12)$$

The bulk modulus and first-order derivative of bulk modulus corresponding to Singh and Kao equation of state is expressed as[22]:

$$K = \frac{V}{V_0} K_0\left[1 + \left(K_0' + 1\right) - \left(K_0' + 1\right)\frac{V}{V_0}\right] \quad (13)$$

$$K' = \frac{K_0}{K}\frac{V}{V_0}\left[-1 - (K_0' + 1) + 2(K_0' + 1)\frac{V}{V_0}\right] \quad (14)$$

Similarly, the expression for volume compression and melting temperature corresponds to Singh and Kao's equation of state can be expressed as:

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$$\frac{V}{V_0} = 1 - \left[\frac{-1 + \sqrt{1 + \frac{2(K_0^{'} + 1)P}{K_0}}}{K_0^{'} + 1} \right] \tag{15}$$

$$T_m(P) = T_0 + (T_{m0} - T_0) \frac{\left(K_0^{'} + 1\right)}{\left(K^{'} + 1\right)} \frac{V}{V_0} \tag{16}$$

2.3. ***Shanker equation of state:*** Considering the Born lattice theory and deriving the volume derivative of the short-range force constant, Shanker obtained an equation of state known as Shanker EOS [23].

$$P = \frac{3K_0 \left(\frac{V}{V_0}\right)^{-\frac{4}{3}}}{\left(3K_0^{'} - 8\right)} \left[\left\{ \left(1 - \frac{1}{t} + \frac{2}{t^2}\right)\left(\exp ty - 1\right) \right\} + \left\{ y\left(1 + y - \frac{2}{t}\right)\exp ty \right\} \right] \tag{17}$$

Where, $y = 1 - \frac{V}{V_0}$ and $t = K_0^{'} - \frac{8}{3}$

The expression for bulk modulus, the first-order derivative of bulk modulus, volume compression, and melting temperature corresponding to Shanker equation of state is expressed as:

$$K = \frac{4}{3}P + K_0 \left(\frac{V}{V_0}\right)^{-\frac{4}{3}} \exp\left\{ \left(K_0^{'} - \frac{8}{3}\right)\left(1 - \frac{V}{V_0}\right) \right\} \tag{18}$$

$$K^{'} = \frac{16}{9}\frac{P}{K} + \left(1 - \frac{4}{3}\frac{P}{K}\right)\left[\left\{ \left(K_0^{'} - \frac{8}{3}\right)\left(\frac{V}{V_0}\right) \right\} + \frac{8}{3} \right] \tag{19}$$

$$\frac{V}{V_0} = 1 + \frac{1 - \sqrt{1 + \frac{2(K_0^{'} + 1)P}{K_0}}}{K_0^{'} + 1} \tag{20}$$

$$T_m(P) = T_0 + (T_{m0} - T_0) \frac{K\left(K_0^{'} + 1\right)}{K_0(K^{'} + 1)} \frac{V}{V_0} \tag{21}$$

2.4. ***Usual-Tait equation of state:*** Usual-Tait proposed a new EOS, known as Usual-Tait EOS, with a slight modification in the Usual-Tait EOS as[24]:

$$P = \frac{K_0}{\left(K_0^{'} + 1\right)} \left[\exp\left\{ \left(K_0^{'} + 1\right)\left(1 - \frac{V}{V_0}\right) \right\} - 1 \right] \tag{22}$$

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The expression for bulk modulus, the first-order derivative of bulk modulus, volume compression, and melting temperature corresponding to the Usual-Tait equation of state is expressed as[25]:

$$K = K_0\left(\frac{V}{V_0}\right)\exp\left\{\left(K_0^{'}+1\right)\left(1-\frac{V}{V_0}\right)\right\} \tag{23}$$

$$K^{'} = \left(K_0^{'}+1\right)\left(\frac{V}{V_0}\right)-1 \tag{24}$$

$$\frac{V}{V_0} = 1-\frac{1}{K_0^{'}+1}\ln\left[\frac{\left\{P\left(K_0^{'}+1\right)+K_0\right\}}{K_0}\right] \tag{25}$$

$$T_m(P) = T_0 + (T_{m0}-T_0)\frac{K\left(K_0^{'}+1\right)}{K_0(K^{'}+1)}\frac{V}{V_0} \tag{26}$$

2.5. Lindemann's Law: Lindemann's law for melting temperature states that the melting temperature of a solid is proportional to the square of the vibrational frequency of its atoms and inversely proportional to the interatomic spacing. When considering pressure dependence, increased pressure typically raises the melting temperature by reducing the interatomic spacing and increasing the vibrational frequency. Lindemann's law [26] for the melting temperature at high pressure is given as:

$$T_m(P) = T_{m0}\left(1+\frac{K_0^{'}P}{K_0}\right)^{\left(\frac{2}{K_0^{'}}\right)\left(\gamma_0-\frac{1}{3}\right)} \tag{27}$$

Where γ_0 is the Gruneisen parameter at zero pressure.

Table 1: Values of input data for alkali halides [26][27]:

Sample	K_0 (GPa)	K_0'	T_{mo}(K)	γ_0
LiF	66.51	5.31	1115	1.83
LiCl	29.68	5.63	887	2.02
LiBr	23.52	5.68	820	2.17
LiI	16.8	4.3	723	2.45
NaF	46.5	5.28	1261	1.72
NaCl	24	5.35	1074	1.84
NaBr	19.9	5.46	1028	1.88
NaI	15.1	5.59	924	1.94

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KF	30.22	5.36	1119	1.71
KCl	17.35	5.48	1049	1.68
KBr	14.64	5.48	1003	1.68
KI	11.51	5.48	959	1.71
RbF	26.68	5.69	1048	1.74
RbCl	15.58	5.62	988	1.73
RbBr	13.24	5.59	955	1.76
RbI	10.49	5.6	915	1.73

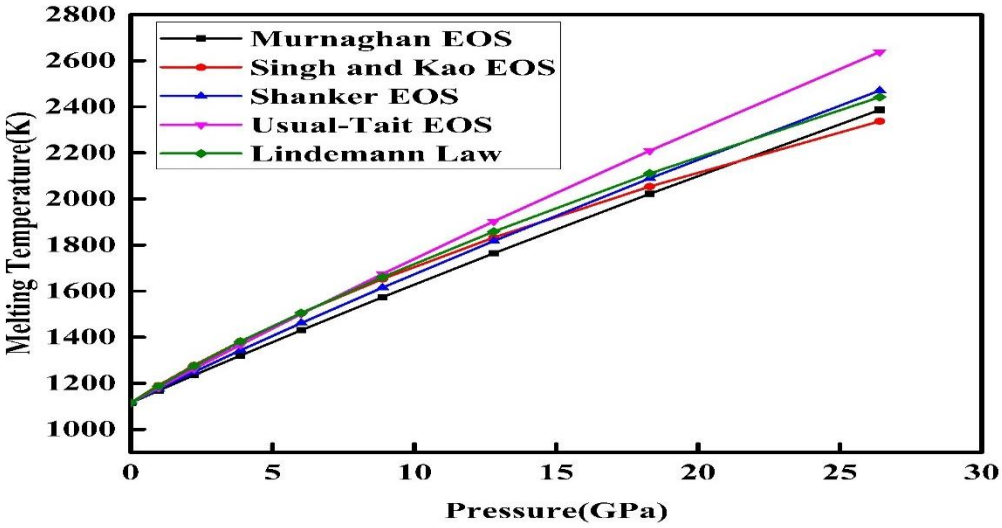

Fig. 1. Pressure-dependent melting temperature of LiF

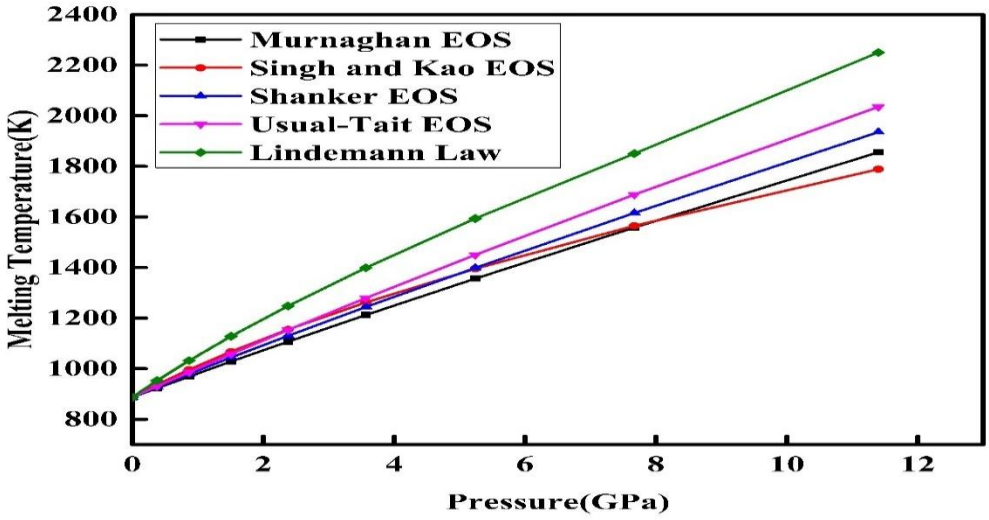

Fig. 2. Pressure-dependent melting temperature of LiCl

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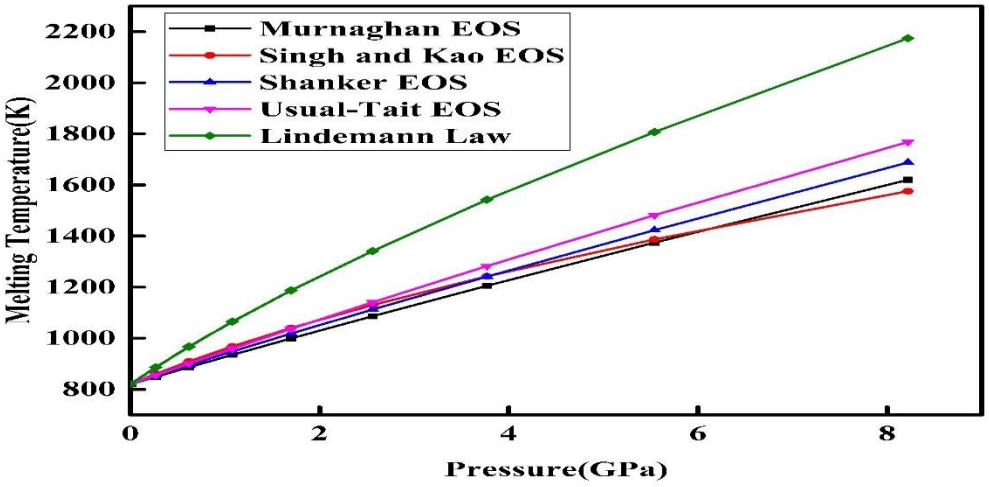

Fig. 3. Pressure-dependent melting temperature of LiBr

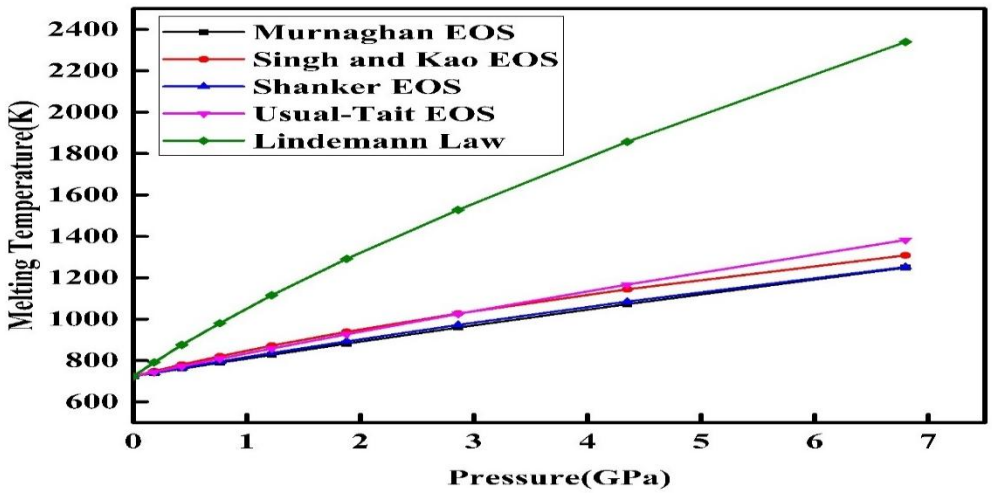

Fig. 4. Pressure-dependent melting temperature of LiI

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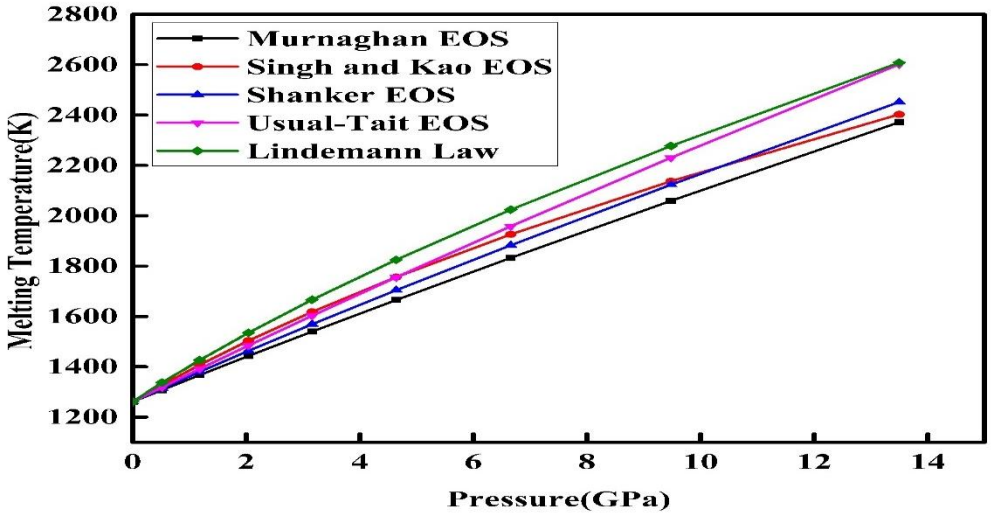

Fig. 5. Pressure-dependent melting temperature of NaF

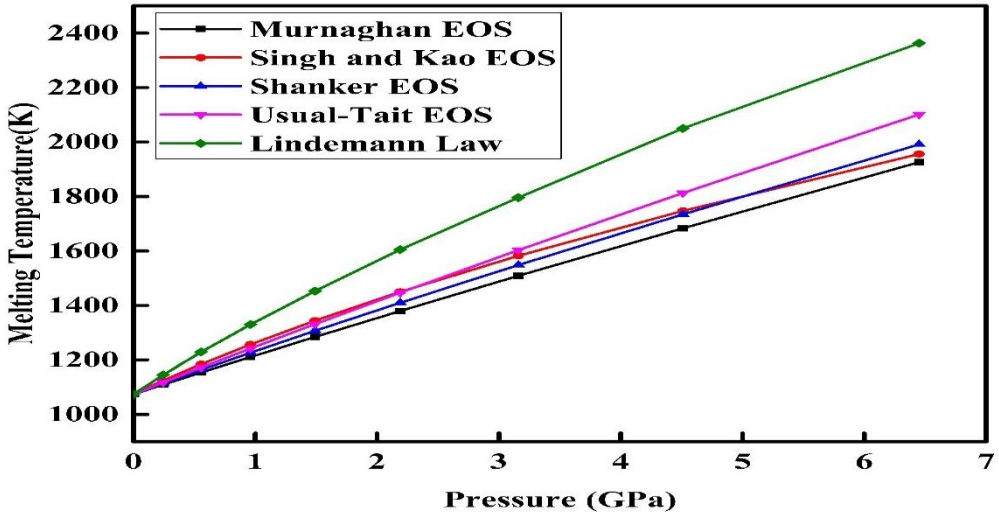

Fig. 6. Pressure-dependent melting temperature of NaCl

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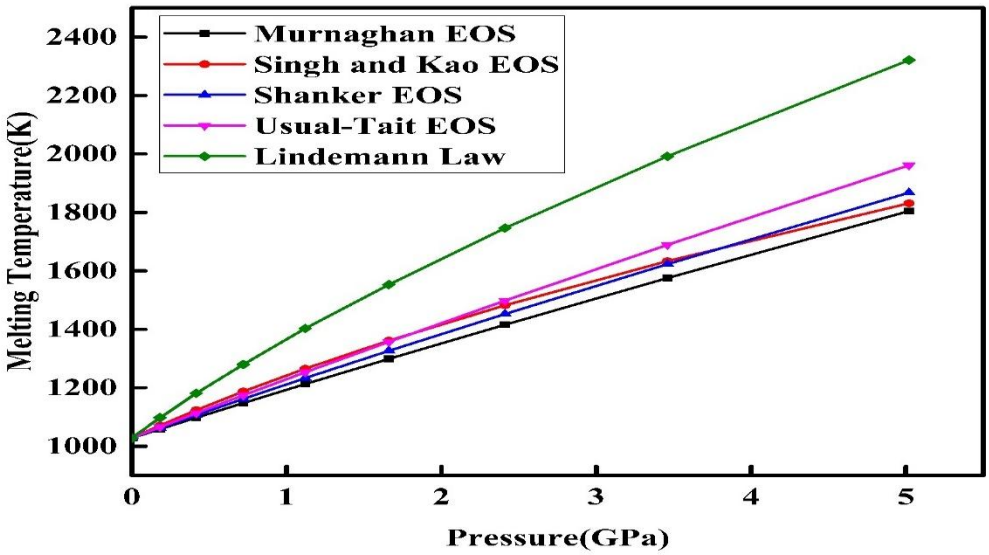

Fig. 7. Pressure-dependent melting temperature of NaBr

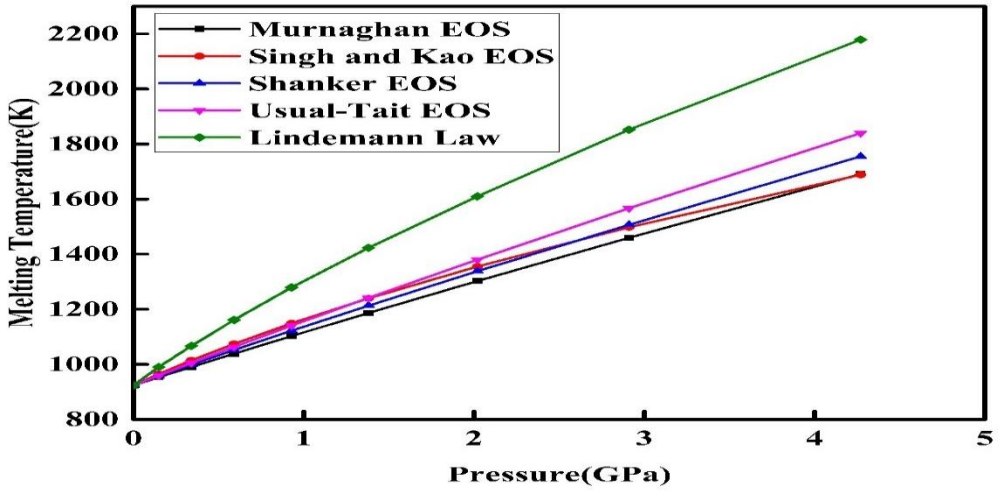

Fig. 8. Pressure-dependent melting temperature of NaI

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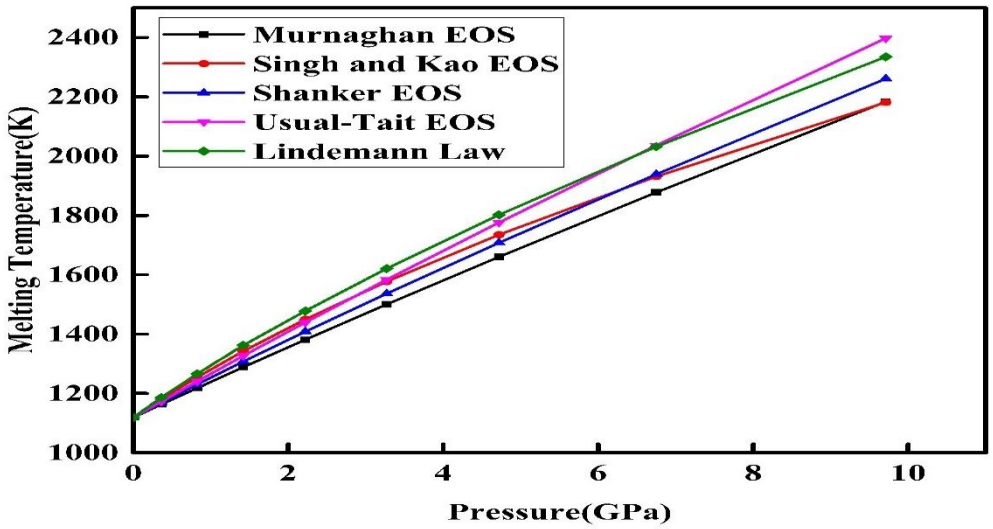

Fig. 9. Pressure-dependent melting temperature of KF

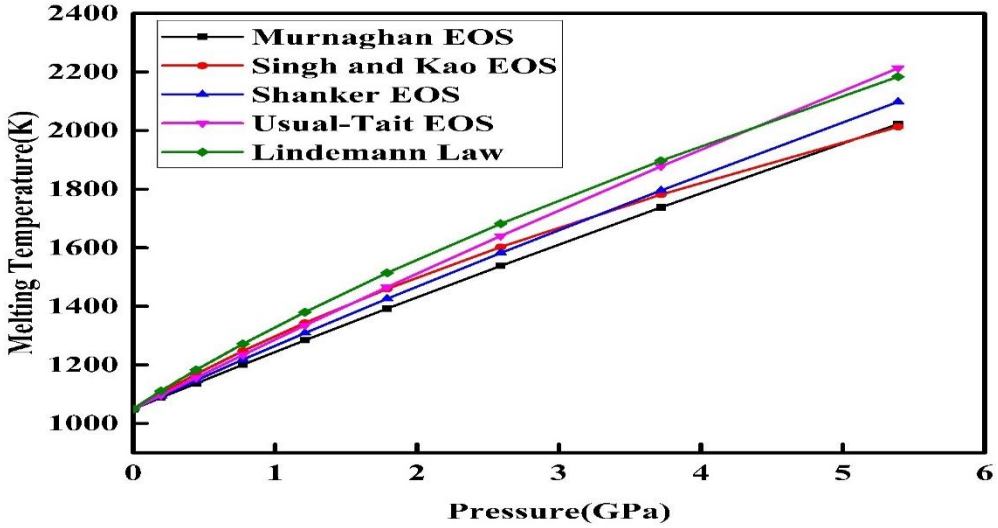

Fig. 10. Pressure-dependent melting temperature of KCl

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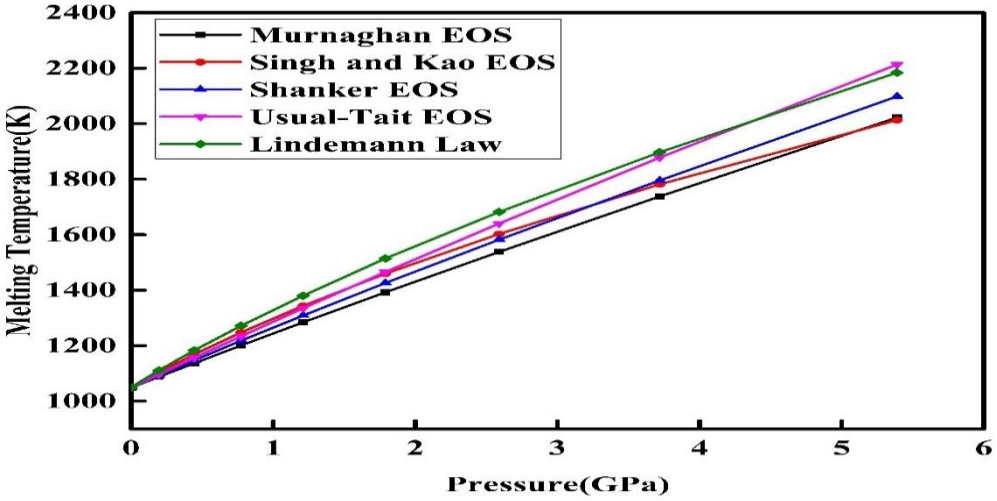

Fig. 11. Pressure-dependent melting temperature of KBr

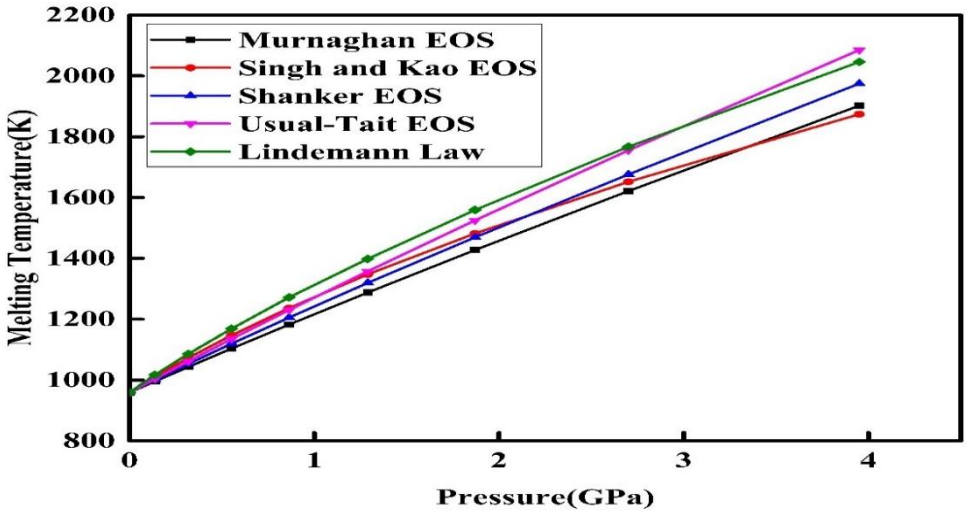

Fig. 12. Pressure-dependent melting temperature of KI

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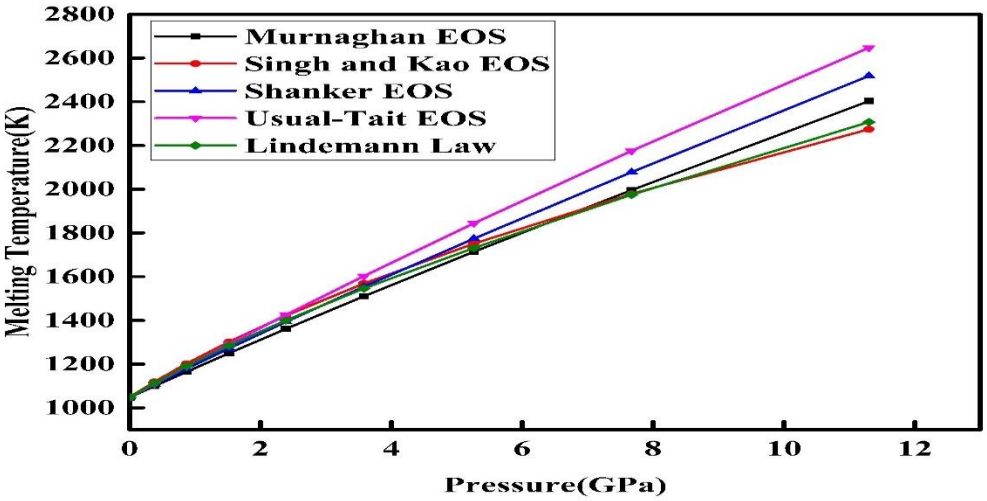

Fig. 13. Pressure-dependent melting temperature of RbF

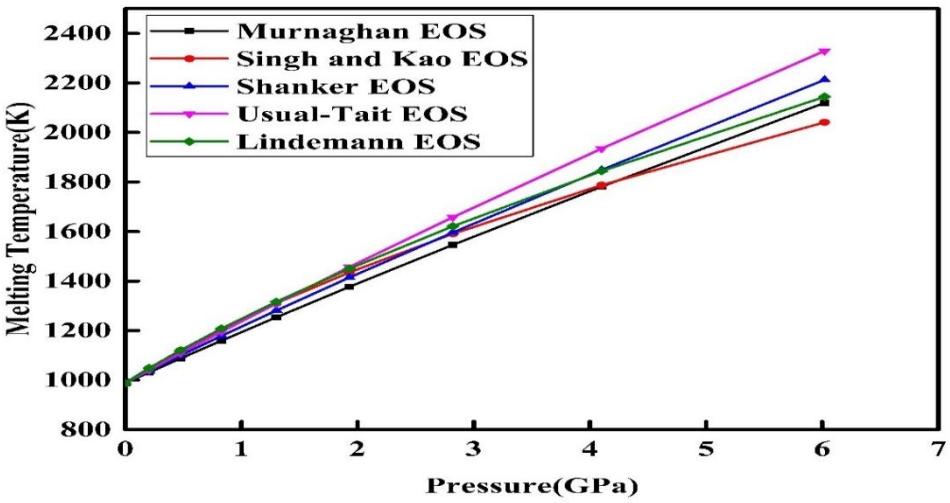

Fig. 14. Pressure-dependent melting temperature of RbCl

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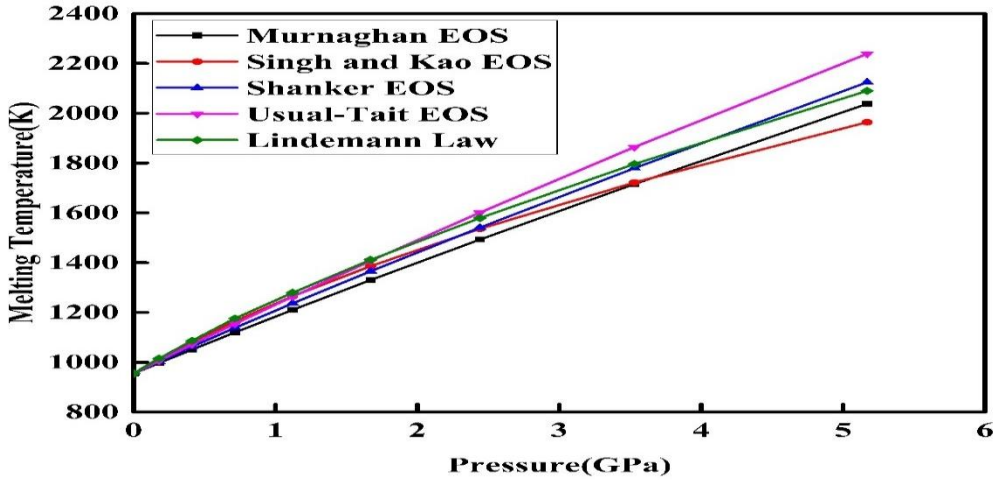

Fig. 15. Pressure-dependent melting temperature of RbBr

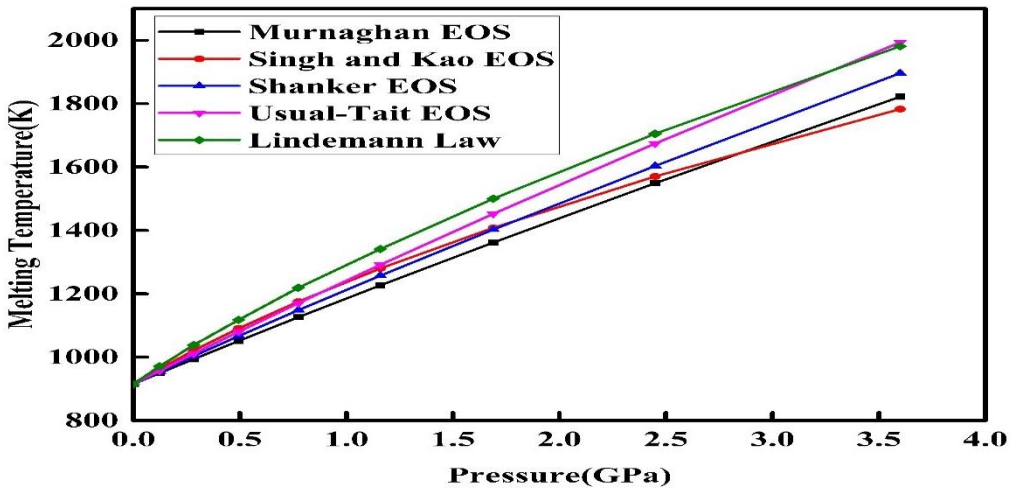

Fig. 16. Pressure-dependent melting temperature of RbI

3. Result and discussion:

The proposed model utilizes three input parameters, B_0, B_0', and T_{m0}, Bulk modulus and its first-order pressure derivative stabilize the melting temperature under high pressure. Table 1 clearly outlines the values of the input parameters for the alkali halides in this study, along with the corresponding references.

The study involved deriving the values of bulk modulus, the first-order pressure derivative of bulk modulus, and the volume compression of alkali halides using various isothermal equations of state, as detailed in section 2. The alkali halides included in the model are LiF, LiCl, LiBr, LiI, NaF, NaCl, NaBr, NaI, KF, KCl, KBr, KI, RbF, RbCl, RbBr, and RbI. The study employed the Murnaghan EOS, Singh and Kao EOS, Shanker EOS, and Usual-Tait EOS to obtain the pressure dependence of volume compression, melting temperature, bulk modulus, and first-order pressure derivatives.

The calculation of bulk modulus, volume compression, first-order pressure derivatives of bulk modulus, and melting temperature for sixteen alkali halides has been carried out using four different equations of state: Murnaghan EOS, Singh and Kao EOS, Shanker EOS, and Usual-Tait EOS. The data indicates that the bulk modulus approaches infinity at extremely high pressures and increases with increasing pressure. The bulk modulus is inversely proportional to the first-order pressure derivative but converges at higher pressures. The Stacey equation of state is valid for both zero-pressure and infinite-pressure conditions [28].

The equation of state describes how the melting temperature and pressure are related to different alkali halides. To better understand this, the results from other state equations are compared with those from Lindemann's law. Curves showing the relationship between the melting temperature and pressure for different alkali halides have been plotted and are displayed in Figure 1-16.

Based on the data analysis, the Usual-Tait EOS generally performs better than other equations of state (EOS) for most alkali halides in terms of efficiency, except for LiF, KBr, RbF, RbCl, and RbBr. For LiF and KBr, the recommended choice is the Shanker EOS, while for RbF, the Singh and Kao EOS provides more precise results. However, the Murnaghan EOS offers poor outcomes due to the invariance of the first-order pressure derivative of the bulk modulus about pressure. It is noteworthy that the alkali halides have a high bulk modulus and a high melting temperature. RbF, Singh, and Kao provided better results than the other EOS for the sample.

High-pressure melting curves of various alkali halides are shown in Figures 1-16. These curves consistently demonstrate that the Usual-Tait equation of state agrees with the melting temperature calculated by the Lindemann law. Therefore, the Usual-Tait EOS is considered the most suitable state equation for accurately predicting alkali halides' melting temperature.

The disparities in calculated results arising from the equation of state and Lindemann's law are primarily due to considerations of compression and assumptions made in deriving the equation of state. Equations of state accounting for higher compression yield more accurate results compared to those based on lower compression. Consequently, as the degree of compression increases, the equation of state provides precise, increasing outcomes.

4. Conclusion:

In our study, we used the Murnaghan EOS, Singh and Kao EOS, Shanker EOS, and Usual-Tait EOS to predict the pressure-dependent melting temperature of alkali halides. These models are valid under high pressure and do not require any adjustable parameters. It has been established that the current equation meets Stacey's requirement for an equation of state [29]. The proposed model also predicts the melting temperature of halides under both low and high pressure. Furthermore, the melting temperature calculated by Lindemann's law matches the computed values by Shanker EOS at different pressures. This research has revealed a link between the bulk modulus and melting temperature. According to the equation of state proposed by Shanker et al., the corresponding melting temperature increases as the bulk modulus increases. [23] is consistent with Lindemann's law, but in the case of LiI and RbF, the models developed by Usual-Tait, Singh, and Kao provide better results than others.

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instilled in me by our parents, which have propelled me to engage in meaningful and impactful research work.

Ethical Approval:
The authors confirm that the manuscript is original and unpublished, and this is a theoretical formulation.

Conflicts of interest:

The authors of this paper declare no known financial interests or personal relationships that could have affected the presented work.

Author's Contribution:

All the authors collaborated to create the research outline. Abhay P Srivastava performed all the necessary calculations and drafted the initial manuscript. Professor B. K. Pandey provided resources and guidance throughout the project. Dr. Anod Kumar Singh, Dr. Reetesh Srivastava, and Harish Chandra Srivastava help us to formatting the manuscript.

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The data supporting this study's findings are available from the corresponding author upon reasonable request.

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