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Research Article

Development of Nano Thermodynamics With its Applications to Different Types of Nanomaterials

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Abstract— Simple thermodynamics is discussed to study the properties of nanomaterials. The method is used to study the size and shape dependence of thermodynamic properties of nanomaterial. We applied it to predict the compression, thermal expansion and their size and shape dependence of different type of nanomaterials. The results are compared with the available experimental data. A good agreement between theory and experiment demonstrates the validity of the theory. It is found that the theory reduces the number of parameters and is applicable to different types of nanomaterials, under varying conditions of size, shape, pressure and temperature. The application to different type of nanomaterials demonstrates the universal nature of the theory. Such simplicity, wide applicability and good agreement with experimental data never seen earlier for nanomaterials.

Keywords — Size, shape, thermodynamics, nanomaterials, equation of state.

1. Introduction

It has been discovered that nanoparticles differ from bulk materials in terms of their behaviour. High surface to volume ratio is responsible for this. Nanoscience and technology are now expanding quickly. The nanotechnology focuses on phenomena and material manipulation at the nano scale. Applications of systems using nanoscale shape and size control are known as nanotechnologies. Grain size, shape, pressure and temperature conditions of nanomaterials are very useful at present. It has been well recognised that properties depend on size, shape, temperature and pressure. The studies of nanomaterials for various conditions are very important for the extension of the material science. Increasing temperature and pressure have many applications and several paths for assembling or phase transformations that constitute a new way for the innovation of new materials.

Effect of size and shape on nanomaterials is well known in the literature [1,2] in addition to the pressure and temperature effects [3]. There are some models that shows the effect of size on the behavior of nanomaterials [4]. While some work on metals and insulators has been recorded, semiconductors have been the focus of the majority of high-pressure research on nanocrystalline materials. Generally speaking, but at higher pressure, it has been discovered that nanocrystals go through the same phase change as their large grain counterparts[5-8]. It has been recognised that equation of state (EOS) and bulk modulus also depend on size of nano materials[9-10]. Jiang et al[11] studied compressibility of nano-structured Fe-Cu experimatally.

Chen et al[8] reported experimental studies for nanocrystalline Fe using diamond anvil cell upto 46GPa and used Birch-Murnaghan (BM) EOS to explain the results. Swamy et al[12] studied the compression behaviour of TiO_2 to 35 GPa using synchrotron X-ray diffraction. Liu et al[13] used diamond anvil cell in situ high pressure Raman scattering and energy dispersive X-ray diffraction to study compression of 3C-SiC and He et al[14] reported the compression of SnO_2 nanocrystals. Marquardt et al[15] reported the experimental studies for MgO. Xiao et al[16] studies compression behaviour of $CsPbBr_3$ using Diamond anvil cell. Rhenium diboride (ReB_2) is a super hard material which can scratch the diamond and may be synthesized under the ambient pressure. Lei et al[17] studied the compression of $Re_{0.52}W_{0.48}B_2$ and ReB_2. Recently, Yue et al[18] reported the high-pressure behaviour of fullerene-like nanoparticles (IF-WS_2). During all these studies BM EOS has been used, which depends on the bulk modulus (B_o) and its first order derivative (B_0') at zero pressure (using the notations of present work).

Due to the wide application of pressure and temperature, several other EOS models have been reported in the literature[20-24]. All these EOS were proposed for bulk material. However, few of them deserve to be used for nanomaterials as discussed by Bhatt and Kumar [25]. Moreover, their application needs the input parameters B_o and B_0'. It should be mentioned that these parameters are not

available for nanomaterials. Their theoretical calculations or experimental measurement for nanomaterials seems to be very difficult. Therefore, it become legitimate and also useful to develop the theory free from the problems as discussed above, using the concept of nanoscience and reduce the number of parameters and also consider the temperature, size and shape effects. However, it is much difficult task in theory. Moreover, we devoted efforts.

2. Theoretical formulation

One of the elastic constants is the bulk modulus. This predicts the compressibility of the material. It is well known that the bulk modulus depends on the temperature as well as pressure, for bulk materials [26]. For nanomaterials the bulk modulus also depends on the size and shape in addition to the pressure and temperature. It becomes much difficult to predict the bulk modulus of nanomaterials under varying conditions of size, shape, pressure and temperature. Recently, Pandey and Kumar [27] reported the theory for this purpose of bulk modulus for nanomaterials, which gives following relation

$$\frac{B_n}{B_b} = \left(1 - \frac{N}{2n}\right)^4 \tag{1}$$

where B_n the bulk modulus of nanomaterial. B_b the bulk modulus of bulk material and N the number of surface atoms, n the total number of atoms in solid.

B_n is defined as

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

Eq. (1) and Eq. (2) gives, (at constant temperature)

$$-\frac{V}{B_b}\left(\frac{dP}{dV}\right) = \left(1 - \frac{N}{2n}\right)^4 \tag{3}$$

Integrating both sides, we get

$$-\frac{1}{B_{0b}}\left(1 - \frac{N}{2n}\right)^{-4}\int_0^P dP = \int_{V_0}^{V}\frac{dV}{V} \tag{4}$$

$$-\frac{1}{B_{0b}}\left(1 - \frac{N}{2n}\right)^{-4} P = ln\left(\frac{V}{V_0}\right) \tag{5}$$

or

$$P = -B_{0b}\, ln\left(\frac{V}{V_0}\right)\left(1 - \frac{N}{2n}\right)^4 \tag{6}$$

Eq. (6) gives the relation of pressure and volume under isothermal condition. An important point is that it contains only one parameter B_{ob}, which corresponds to the bulk state of the concerned material and readily available in the literature. Thus, Eq. (6) reduces the number of parameters.

It has been discussed by Bhatt and Kumar [2] that N/2n depends on size and shape. These values for different shapes have been compiled in Table1. Thus, we get the EOS for different shape of nanomaterials as given below:

Film:

$$P = -B_{0b}\ln\left(\frac{V}{V_0}\right)\left(1 - \frac{0.666d}{h}\right)^4 \tag{7}$$

Dodecahedral:

$$P = -B_{0b}\, ln\left(\frac{V}{V_0}\right)\left(1 - \frac{0.898d}{a}\right)^4 \tag{8}$$

Icosahedral:

$$P = -B_{0b}\ln\left(\frac{V}{V_0}\right)\left(1 - \frac{1.323d}{a}\right)^4 \tag{9}$$

Wire:

$$P = -B_{0b}\ln\left(\frac{V}{V_0}\right)\left(1 - \frac{1.333d}{L}\right)^4 \tag{10}$$

Spherical:

$$P = -B_{0b}\ln\left(\frac{V}{V_0}\right)\left(1 - \frac{2d}{D}\right)^4 \tag{11}$$

Hexahedral:

$$P = -B_{0b}\ln\left(\frac{V}{V_0}\right)\left(1 - \frac{2d}{a}\right)^4 \tag{12}$$

Octahedral:

$$P = -B_{0b}\ln\left(\frac{V}{V_0}\right)\left(1 - \frac{2.449d}{a}\right)^4 \tag{13}$$

Tetrahedral:

$$P = -B_{0b}\ln\left(\frac{V}{V_0}\right)\left(1 - \frac{4.898d}{a}\right)^4 \tag{14}$$

We used Eqs (7-14) for the study of shape effect on nanomaterials.

3. Temperature Effect

To include the temperature effect in Eq. (6), we may develop the as follows. The thermal pressure reads as[28]

$$\left(\frac{\partial P_{Th}}{\partial T}\right)_V = \alpha_{0n}B_{0n} \tag{15}$$

α_{0n} is volume thermal expansion coefficient and Eq. (15) can be integrated as follows[28]

$$P_{Th} = \int_{T_0}^{T}\alpha_{0n}B_{0n}\,dT$$

or

$$P_{Th} = \alpha_{0n}B_{0n}(T - T_0) \tag{16}$$

Now including the temperature effect, in Eq. (6) using (16), we get the following relation

$$P = -B_{ob}\ln\left(\frac{V}{V_0}\right)\left(1 - \frac{N}{2n}\right)^4 + \alpha_{0n}B_{0n}(T - T_0) \tag{17}$$

Eq. (17) may be regarded as EOS for nanomaterials. At P=0, using the relation $\alpha_{0b}B_{0b} = \alpha_{0n}B_{0n}$ as discussed earlier [29], we get the relation

$$\frac{V}{V_0} = exp\left[\alpha_{0b}\left(1 - \frac{N}{2n}\right)^{-4}(T - T_0)\right] \tag{18}$$

Now, putting the values of N/2n as given in Table 1, we formed the formulation of different shapes for thermal expansion and used in the present paper.

4. Results And Discussion

We have developed a simple theory to understand the effect of size, shape, pressure and temperature on nanomaterials. The theory gives Eq. (6) for the size and shape dependence of compression behaviour. To test the validity of model, we selected different types of nanomaterials. The selection of these materials is based on the condition that the experimental information is available so that the model predictions may be compared. The input data [8,11-19] are compiled in Table 1. We used Eq. (6) to compute pressure dependence of V/V_0 for different nano materials. These results are summarized in Figures 1-10 with the experimental data [8,11-19]. Very good agreement between theoretical predictions and experimental data are found. This demonstrates the validity of the theory developed in present paper. During high pressure studies of materials, it has been found[24] that the maximum deviations with theory and experiments occur at highest pressures. We have therefore calculated the percentage deviations at highest pressures, which have been reported in Table 1. This clearly demonstrate the validity of the model proposed during present study. Eq. (6) may also be used to study the shape effect using the values of N/2n from Table 1 which gives the Eq. (7-14). Using these equations, we have computed the compression of nanomaterials for several shapes. The results obtain are reported in Figure 11-12. This shows that compression behaviour is affected by shape also.

It should be mentioned that Eq. (6) has been derived at constant temperature and therefore may be regarded as isothermal EOS. To make it complete, we have added thermal pressure terms, which gives Eq. (17). Now Eq.(17) is the complete EOS for nanomaterials. An important point is that it includes the effect of size and shape, an important aspect of nanoscience. A review of literature shows that a sufficient information is available regarding thermal expansion of nanomaterials. At P=0, Eq. (17) may be rewritten as Eq. (18). We used our Eq. (18) to predict V/V_0 at different temperatures. The results are reported in Figures 13-17. A good agreement between theory and experiment [31-35] further reflects the suitability of the model proposed. The model predicts that V/V_0 depends linearly on temperature. The linear behaviour, have also been reported experimentally for nanomaterials[30]. Thus, the model predictions are also consistent with the experimental observations of Lu et al [30] for nanomaterials.

Eq. (18) has been further used to understand the effect of shape on thermal expansion of nanomaterials. This depends on the values of N/2n. By using these values as compiled in Table 1, we get the corresponding relations. The computed values of shape dependent of thermal expansion are reported in Figure 18-19. It is observed that V/V0 is maximum for tetrahedral shape and minimum for film. For other shapes the values lie in between these two sets. Thus, thermal expansion depends on shape also in addition to size, we are reporting these theoretical results in the absence of experimental data for the benefit of researchers engaged in the thermal properties of nanomaterials.

Table 1. Input data used in the present work[2,8,11-19].

Material	B_{0b} (GPa)	Pressure (GPa)	P. D.	Shape	N/2n	Material	d(nm)	α_{0b} (10^{-5} K^{-1})
Fe-Cu (14 nm)	151	7	0.11	Film	0.666 d/h	Se	0.437	9.45
Fe (10 nm)	179.4	42	0.59	Dodecahedral	0.898 d/a	ZnO	0.368	1.05
TiO_2 (40nm)	180	16	0.56	Icosahedral	1.323 d/a	Al	0.286	7.0
3C-SiC (30 nm)	260	21	0.31	Wire	1.333 d/L	Ni	0.248	3.3
SnO_2 (14nm)	225	40	0.13	Spherical	2d/D	Fullerene	0.220	1.55
MgO (20nm)	177	29	2.1	Hexahedral	2d/a			
$CsPbBr_3$ (11.7nm)	20.9	1	0.32	Octahedral	2.449 d/a			
$Re_{0.52}W_{0.48}B_2$ (120 nm)	314	53	1.2	Tetrahedral	4.898 d/a			
ReB_2 (50nm)	343	34	1.1					
IF-WS_2 (95nm)	56.7	14	2.5					

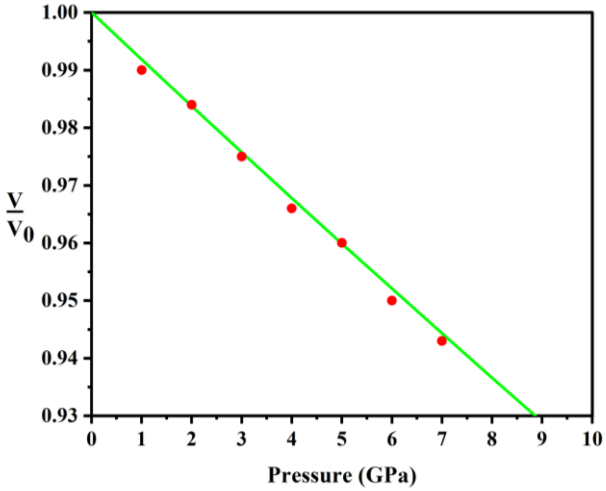

Figure 1. Effect of pressure on V/V_0 for Fe-Cu, – represents present work (Eq.6) and ● experimental data [11].

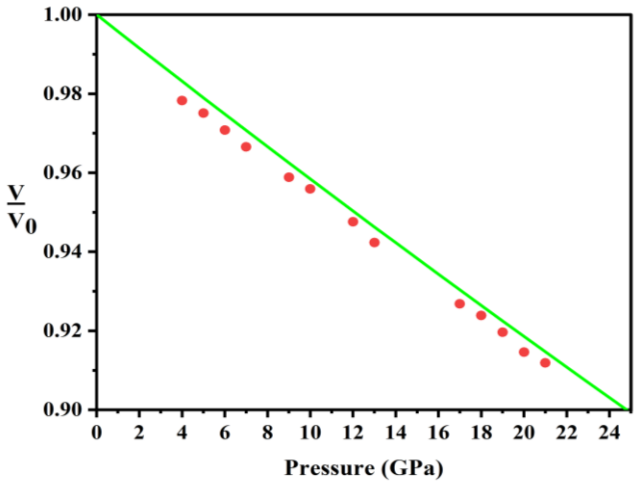

Figure 4. Effect of pressure on V/V_0 for 3C-SiC, – represents present work (Eq.6)and ● experimental data [13].

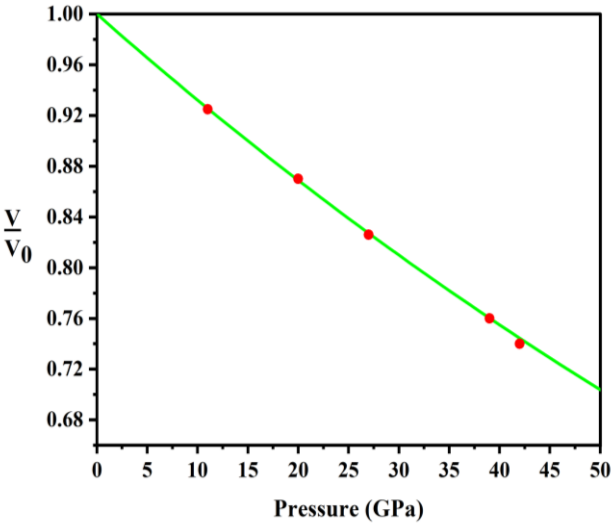

Figure 2. Effect of pressure on V/V_0 for Fe, – represents present work (Eq.6) and ● experimental data [8].

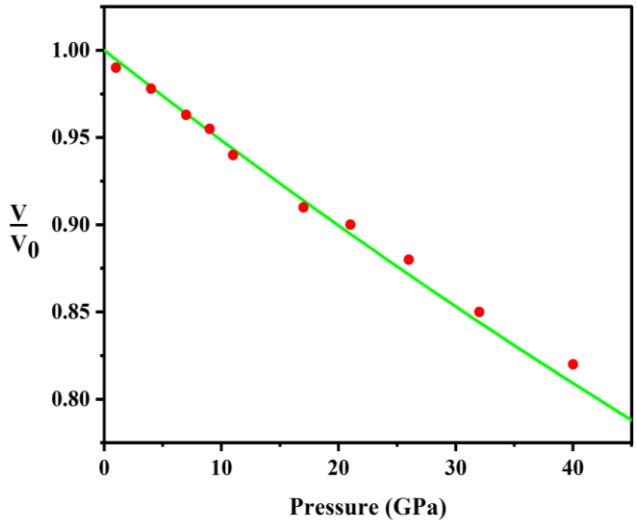

Figure 5. Effect of pressure on V/V_0 for SnO_2, – represents present work (Eq.6) and ● experimental data [14].

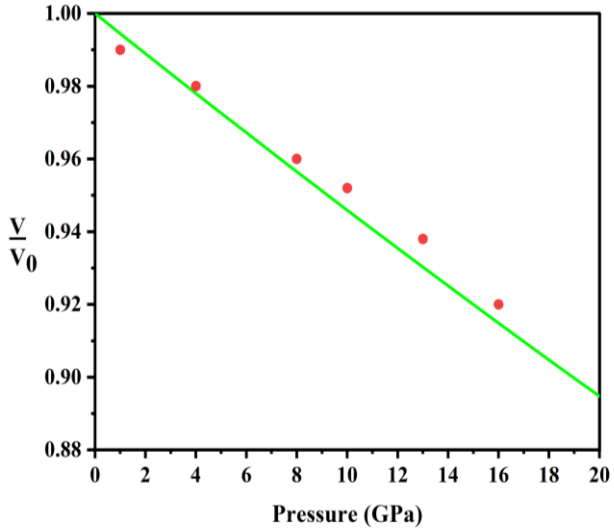

Figure 3. Effect of pressure on V/V_0 for TiO_2, – represents present work (Eq.6) and ● experimental data [12].

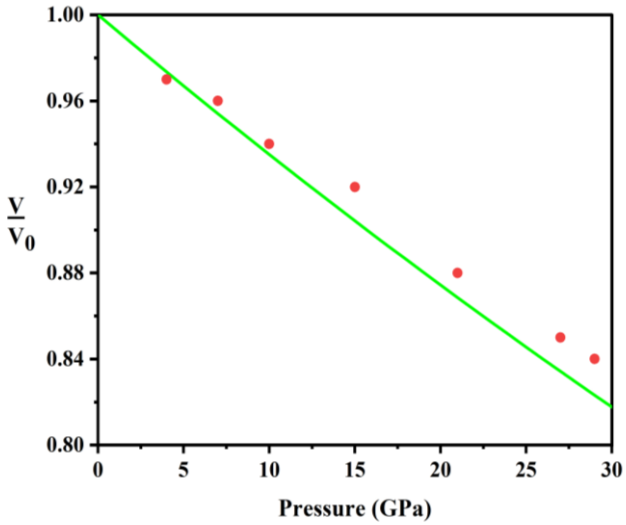

Figure 6. Effect of pressure on V/V_0 for MgO, – represents present work (Eq.6) and ● experimental data [15].

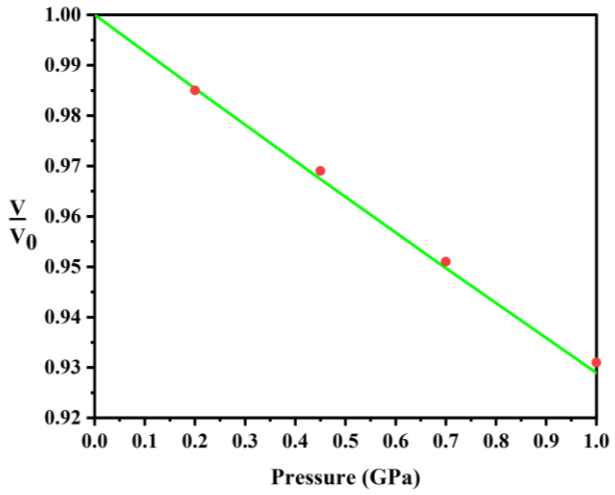

Figure 7. Effect of pressure on V/V_0 for $CsPbBr_3$, – represents present work (Eq.6) and ● experimental data [16].

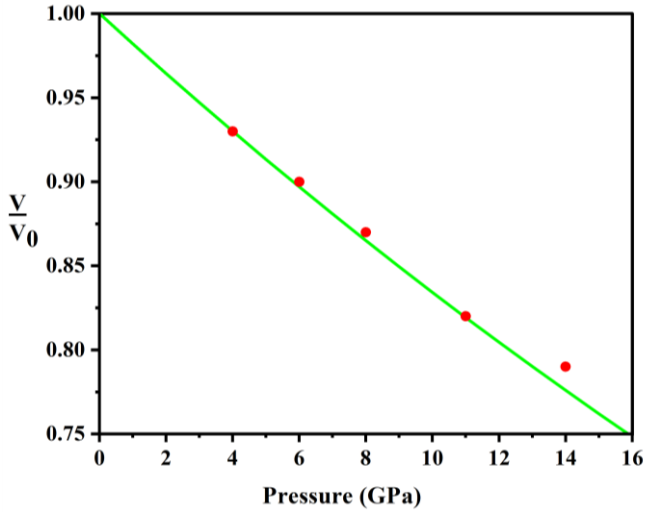

Figure 10. Effect of pressure on V/V0 for IF-WS_2, – represents present work (Eq.6) and ● experimental data [18].

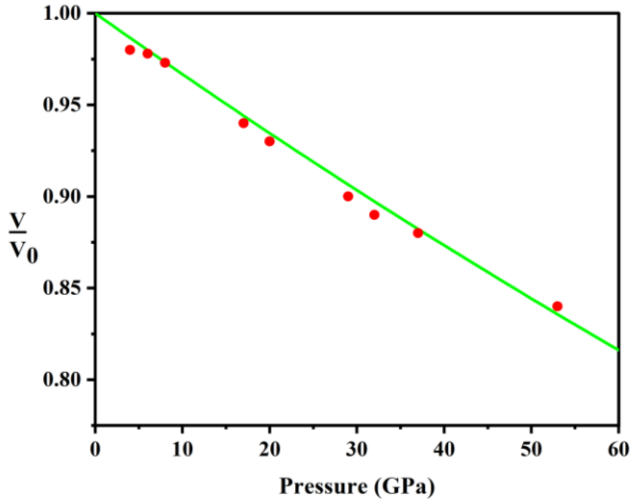

Figure 8. Effect of pressure on V/V_0 for $Re_{0.52}W_{0.48}B_2$, – represents present work (Eq.6) and ● experimental data [17].

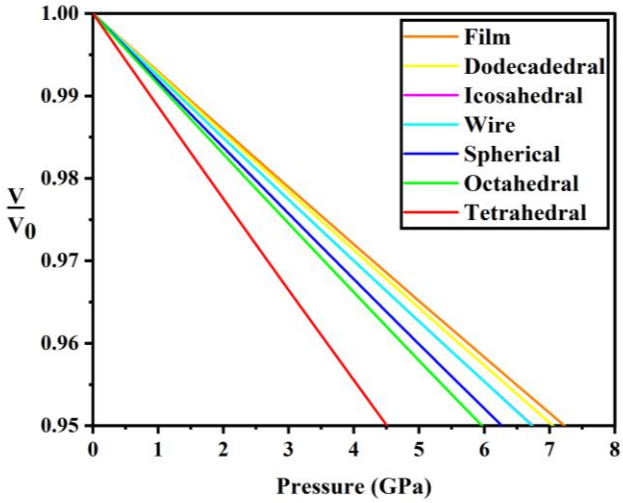

Figure 11. Effect of pressure on V/V_0 for Fe-Cu using Eq. (7-14) including shape effect.

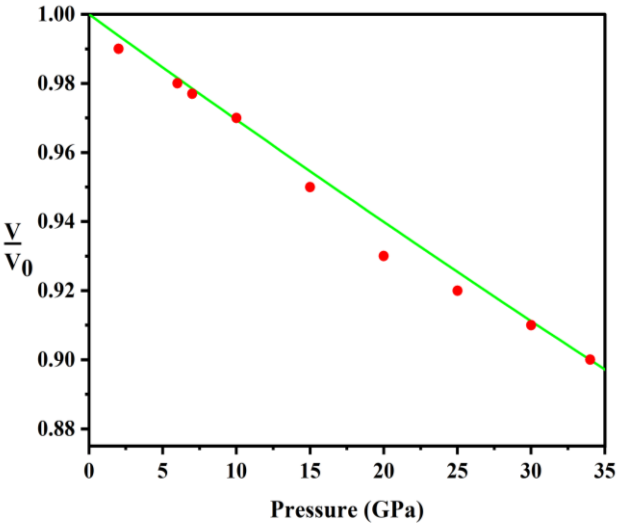

Figure 9. Effect of pressure on V/V_0 for ReB_2, – represents present work (Eq.6) and ● experimental data [17].

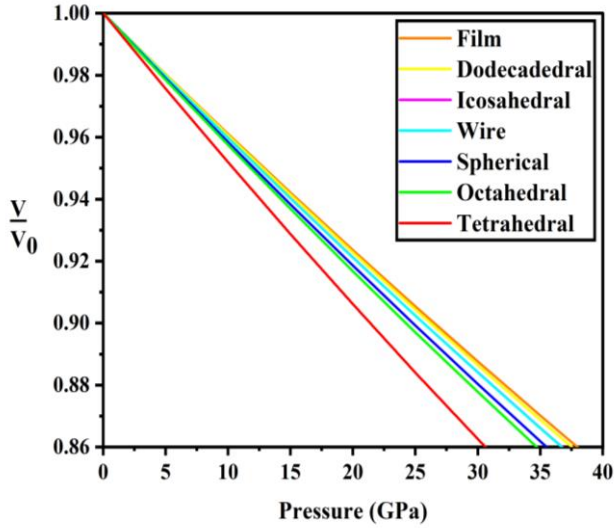

Figure 12. Effect of pressure on V/V_0 for 3C-SiC using Eq. (7-14) including shape effect.

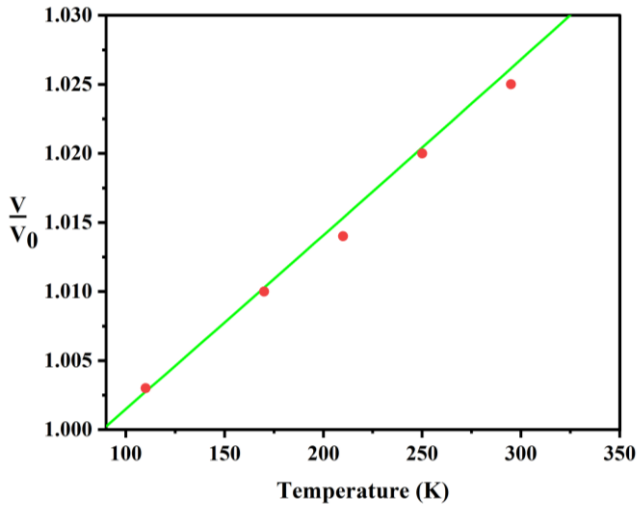

Figure 13. Effect of temperature on V/V_0 for Se, – represents present work Eq. (18) and ● experimental data [31].

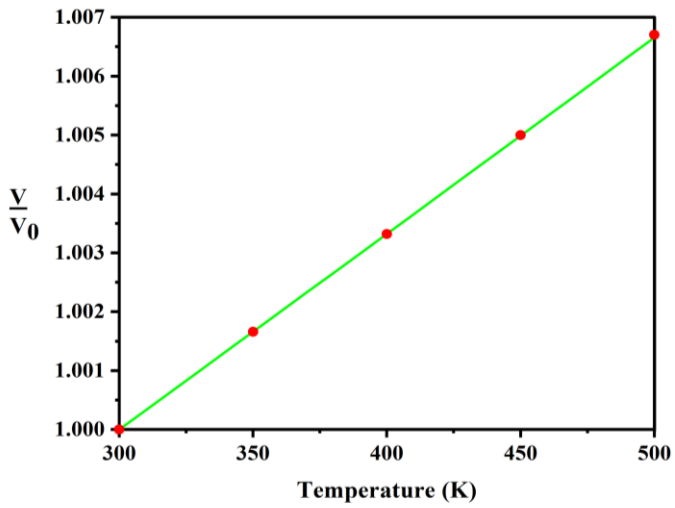

Figure 16. Effect of temperature on V/V_0 for Ni, – represents present work Eq. (18) and ● Experimental data [34].

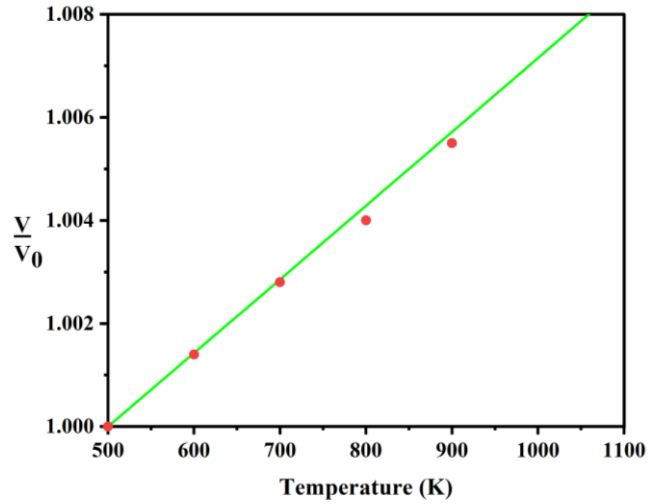

Figure 14. Effect of temperature on V/V_0 for ZnO, – represents present work Eq. (18) and ● experimental data [32].

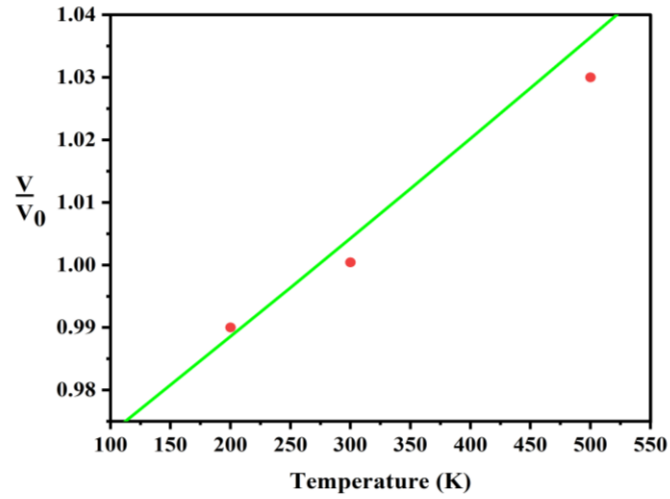

Figure 17. Effect of temperature on V/V_0 for Fullerene, – represents present work Eq. (18) and ● experimental data [35].

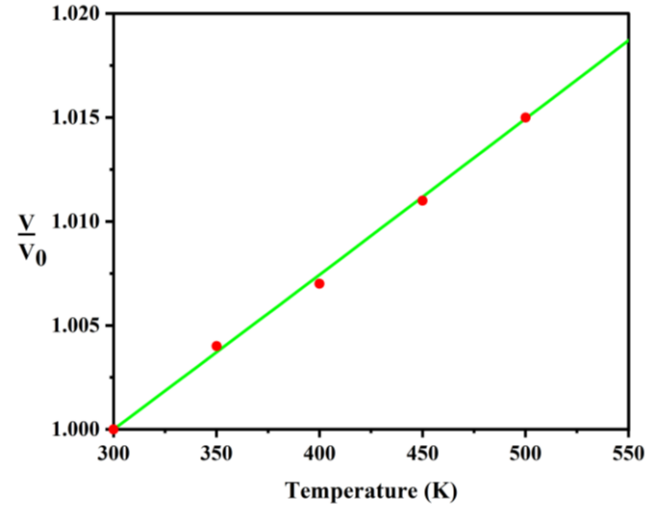

Figure 15. Effect of temperature on V/V_0 for Al, – represents present work Eq. (18) and ● experimental data [33].

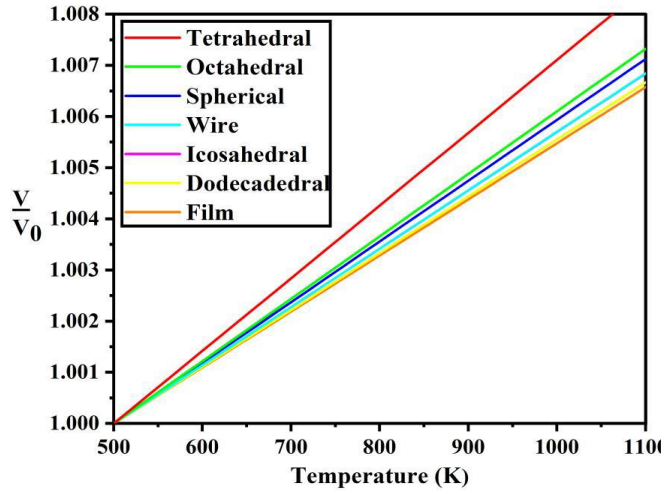

Figure 18. Thermal expansion of ZnO using Eq. (18) including shape effect

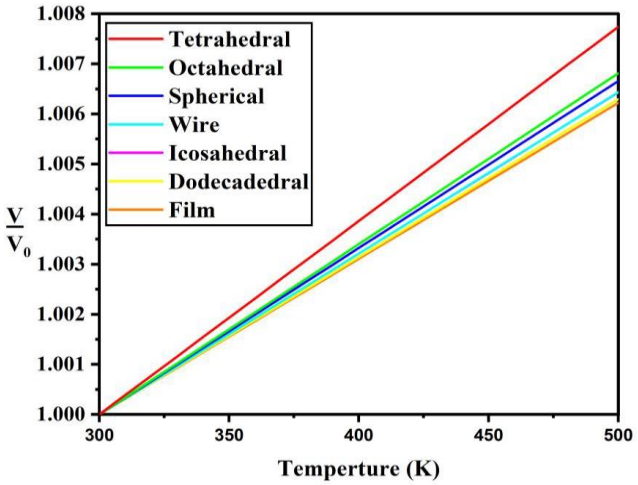

Figure 19. Thermal expansion of Ni using Eq. (18) including shape effect.

4. Conclusion

We have developed the thermodynamics formulation for nanomaterials. This gives the isothermal EOS with single parameter (bulk state). The model has been used to study the size and shape dependence of the compression and thermal expansion of nanomaterial. The present thermodynamics reduces the number of parameters and gives very good agreement with the available experimental data. Some results have also been reported in the absence of experimental data for the benefit of researchers engaged in the study of the pressure, temperature, size and shape behaviour of nanomaterials.

Data availability statement
All data generated or analysed during this study are included in this article. We
have mentioned the source of data in reference.

Conflict of interest statement
The authors declare that they have no known competing financial interest or
personal relationship that could have appeared to influence the work reported in
this paper.

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Author contribution statement
Both the authors (Priya Paneru and Munish Kumar) made their contributions to
the conception or design of the work and analysed properly. We drafted the
work and revised it carefully. We approve the version to be published and agree
to be accountable for all aspects of the work in ensuring that questions related to
the accuracy or integrity of any part of the work are appropriately investigated
and resolved.

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