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**Research Article**

# Pressure-induced valence changes of cerium in some cerium-based compounds

Md Shafiqul Islam, Md. Tarek Hossain and Abdul Hannan*

*Department of Physics, Shahjalal University of Science and Technology, Bangladesh*

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**ABSTRACT**

In the present work, we investigate how pressure affects the valence change of cerium in cerium-based compounds Ce*X* (*X*= O, S, Te, P, As, Sb, and Bi). The valence of cerium at a certain high pressure is estimated by comparing the experimentally observed unit cell volume and the calculated unit cell volume of stable trivalent cerium ion in the compound. The change in unit cell volume with pressure is estimated by employing the Birch equation of state. It is found that the valence of Ce in the Ce*X* is significantly influenced by both the applied pressure as well as the chemical environment around the Ce ion.

## Introduction

Appling hydrostatic pressure on materials can change the properties of the materials. The extra-nuclear electronic states, especially the electronic states of localized *f* electrons in the compounds are responsible for altering the properties. The importance of high pressure work is increasing daily as materials exhibit variety of effects under high pressure. Application of high pressure on compounds produces numerous effects, namely, structural phase transition (Léger, 1993), uncommon resistivity (Okayama et al., 1992), the complicated structure of the magnetic phase (Hannan et al., 2002; Osakabe et al., 2002), change of Kondo effect (Hayashi et al., 2016), pressure-induced valence transition (Hossain et al., 2015), etc. In recent work, Ubukata et al., (2022) investigated the structural change of BaHCl at high-pressure up to 30 GPa using an in-situ high-intensity (synchrotron) X-ray diffraction. They reported that the tetragonal PbFCl-type structure (*P*4/*nmm*) of BaHCl changes into an orthorhombic structure at around 29 GPa.

In this work, the valence alterations of Ce in the two series of compounds, cerium monochalcogenides Ce*X* (*X*=O, S, and Te) and cerium monopnictides Ce*X* (*X*=P, As, Sb and Bi), have been calculated with pressure. Vedel et al. (1986), Léger et al. (1987), and Léger (1993) reported in detail the experimentally observed unit cell volume ($V$) with pressure ($P$) and also the structural phase transitions of these materials. They observed an unusual trend in the $P-V$ curves of these compounds in the high pressure up to 25 GPa. The pressure-volume relationships of the compounds (Léger, 1993) may be reproduced by adopting the Birch equation of state (Birch, 1947; Mito et al., 2007) using appropriate parameters in the equation. This reproduction is necessary to estimate of the valence change with the pressure. A part of this work has already been published (Hossain et al., 2015; Hossain and Hannan, 2017). This paper is dedicated to presenting the more precise calculated results of valence change with pressure for more compounds.

Let us focus on the experimentally observed (Léger, 1993) $P-V$ behavior of Ce*X* (*X*=O, S, and Te). The volume of the cerium oxide (CeO) decreases acutely with the applied pressure of up to 3 GPa and then falls slowly with a further increase in pressure. The NaCl-type structure of this compound remains

*Corresponding author: <ahannan-phy@sust.edu>

unchanged up to the pressure of 25 GPa. The volume drops gradually with pressure for cerium sulphide (CeS) and maintains a NaCl-type structure up to 27 GPa. The volume of CeTe decreases gradually up to 8 GPa, and afterward a structural phase transition occurs from NaCl to CsCl-type.

Among the series Ce*X* (*X*=P, As, Sb and Bi), the compound CeP shows a continuous decrease of volume with pressure, but at around 10 GPa, a large isostructural sudden transition is observed and then continues its continuous volume decrement with further increase of pressure up to about 20 GPa (Léger, 1993; Vedel et al., 1987). This isostructural transition arises due to the valence change (transition) of Ce ions with pressure, as Svane et al. (2001) reported in their theoretical work. Above 20 GPa, this compound shows a structural transition from NaCl to a CsCl-type layout. The volume of CeAs also decreases continuously with pressure until the pressure of 18 GPa is reached, and then the structural transition from NaCl to CsCl-type is occurred at around 18 GPa. CeSb shows a first-order structural conversion from NaCl-type to body central tetragonal structure occurs at 11 GPa. CeBi, on the other hand, shows the structural conversion from NaCl to coexisted CsCl-type and body central tetragonal structures with equal proportion at 13GPa.

Other reported phenomena of these compounds are as follows. Hannan et al., (2000) reported a notable decrease in the crystal lattice of CeSb below a certain temperature $T_L$ at high pressure in the paramagnetic area. This temperature is approximately equal to the resistivity abnormality temperature $T_R$ that appeared in the work of Okayama et al. (1992). Chattopadhyay *et al*. (1994) and Osakabe et al. (2002) reported the complicated magnetic pressure ($P$)−temperature ($T$) phase diagram in CeSb at low temperatures. The magnetic *P-T* phase diagram of CeP (Hannan et al., 2002) is more complicated than that of CeSb. The pressure-dependent electrical resistivity ($\rho$) of CeP is more anomalous than CeAs (Okayama et al., 1992). At pressures above 3 GPa applied on CeSb, an extraordinary rise of $\rho$ while decreasing the temperature is obtained around 30 K (Okayama et al., 1992). It is seen that the peak resistivity increases with the increase of pressure, and it becomes nearly 23 times large at 7 GPa compared with that at ambient pressure. Additionally, CeBi exhibits a sharp peak of $\rho$ at around 30 K coupled with the magnetic phase. Above 3 GPa, this peak converts into a hump and disappears gradually with the increase of pressure.

Hayashi et al. (2016) studied the pressure dependences of various parameters, such as the antiferromagnetic ordering temperature $T_N$, crystal-field splitting, and the ln$T$ anomaly of the Kondo effect in CeS, CeSe, and CeTe. They studied those from the measurement of electrical resistivity under various constant high pressures up to 8 GPa. It is observed that the ln$T$ term in the temperature dependence of $\rho$ in each compound generally increases with the rise of pressure. This result indicates that *c–f* hybridization is increased by applying pressure. The authors discovered two ln $T$ regions in these compounds. The first one is at high temperatures, where both $\Gamma_7$ and $\Gamma_8$ states are involved, which reflects crystal electric field (CEF) splitting, and the second one is at low temperatures, where the $\Gamma_7$ ground state has mainly appeared. From the ln$T$ term in the low-temperature region, Hayashi et al. (2016) found that the *c-f* hybridization strength, $J_{cf}$ is largest in CeS and smallest in CeTe.

Svane et al. (2001) discussed the pressure-induced valence transitions in cerium monopinictides and monochalcogenides associated with isostructural or structural phase transition. De and Chatterjee (1989) studied this electronic band structure of CeS at high pressure using a self-consistent linear augmented plane-wave method in the local-density approximation to assess the change in valence with pressure.

Svane et al. (1999) also studied the electronic structure of CeS, CeSe, and CeTe using the self-interaction corrected local spin-density approximation. They found that the pressure-induced phase transitions are responsible for the delocalization of the *f* electrons, *i. e.*, for the valence change of Ce atoms from trivalent towards tetravalent. So far, we know, work on the

systematic change of valence of cerium in cerium-based compounds in a wide pressure range has yet to be reported except in our previous work, where we reported for several compounds.

To estimate the valence change of Ce of the compounds Ce$X$ ($X$=O, S, Te, P, As, Sb, and Bi) with pressure, we performed the following tasks. The experimentally observed pressure ($P$) −volume ($V$) relationships of those compounds were reproduced by adopting the Birch equation of state (Birch, 1947; Mito et al., 2007) with the bulk modulus ($B_0$) and its first pressure derivative ($B^*$) as fitting parameters. The $P-V$ relationship of those compounds for stable trivalent cerium ions has also been calculated using the same Birch equation but with appropriate bulk modulus and its first pressure derivative. The effect of pressure on the valence of cerium of those compounds has been realized clearly.

## Calculation of relative volume

### Experimental relative volume

Experimental $P-V$ curves (Léger, 1993) of Ce$X$ ($X$=O, S, Te, P, As, Sb and Bi) can be fitted (reproduced) well (or approximately) adopting the Birch equation (Birch, 1947; Mito et al., 2007) as written below:

$$P\left(\frac{V}{V_0}\right)=\frac{3}{2}B_0\left(\left(\frac{V}{V_0}\right)^{-\frac{7}{3}}-\left(\frac{V}{V_0}\right)^{-\frac{5}{3}}\right)\left[1-\frac{3}{4}(B^*-4)\left(1-(V/V_0)^{-2/3}\right)\right] \quad \text{..... (1)}$$

Here, $V/V_0$, $B_0$, and $B^*$ are the relative volume, the Bulk modulus and the pressure derivative of the Bulk modulus, respectively. $B_0$ and $B^*$ are used as fitting parameters during the reproduction of relative volume at each high pressure. IGOR Pro software, Version 8.0 has been used for all calculations to reproduce experimental data.

### Relative volume of CeO

The reproduced experimental $V/V_0$ against $P$ for CeO is shown in Fig. 1 by filled circles. The experimental data has been reproduced perfectly in the pressure range 0 to 3 GPa, utilizing the parameters $B_0=30$ GPa and $B^*=5$. $B_0$ and $B^*$ in the pressure range of about 3 to 23.6 GPa are taken at 1.58 GPa and 679, respectively. It is seen that the reproduction of $V/V_0$ in the pressure range of about 3 to 6 GPa is not consistent with the experimental data of Léger (1993).

### Relative volume of CeS and CeTe

Fig. 2 shows the variation of the reproduced experimental relative volume of CeS with pressure. The experimental data of Léger (1993) has been reproduced using 83.5 GPa and 2.2 for $B_0$ and $B^*$, respectively.

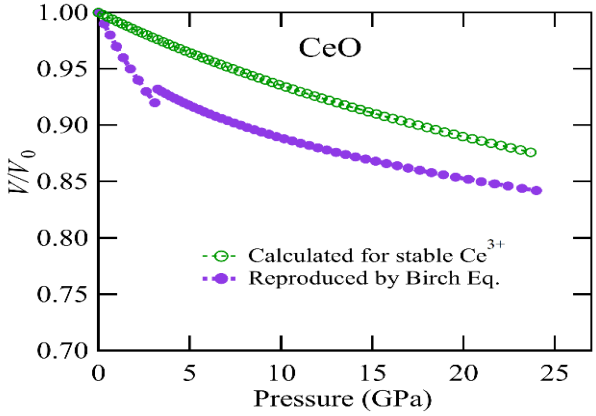


**Fig. 1. Relative volume versus pressure curves for CeO. Filled circles represent experimental data (Léger, 1993) reproduced by Birch equation and open circles represent the data for stable $Ce^{3+}$ ion.**

The variation of the reproduced experimental relative volume of CeTe with pressure is also shown in Fig. 2. The Birch equation has exactly reproduced the experimental data with the parameters $B_0=58$ GPa and $B^*=8$.

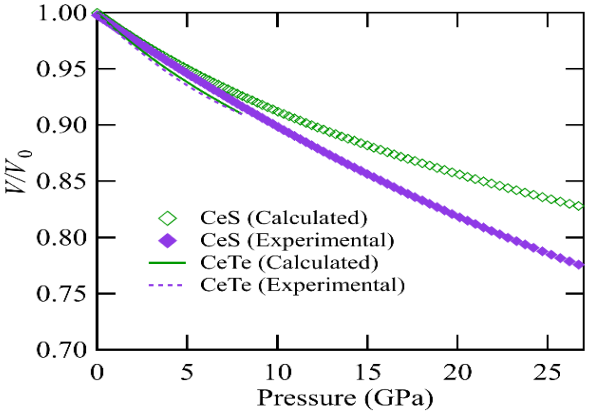


**Fig. 2. Reproduced (experimental) relative volume versus pressure curves for CeS and CeTe (solid diamonds and dashed line) and their estimated curves (open diamonds and solid line) for the existence of stable $Ce^{3+}$ ions in the materials.**

### Relative volume of CeP and CeAs

Fig. 3 shows the reproduced (experimental) relative volumes with pressure for CeP and CeAs. In the case of CeP, the exact reproduction has been attained using the parameters ($B_0$ and $B^*$) 64 GPa and 3 in all pressures between 0 and 9 GPa. The parameters $B_0$ and $B^*$ are chosen to be 30 GPa and 9.2, respectively, to reproduce the experimental data in the pressure from about 9 to 22 GPa. For CeAs, the experimental data has been accurately reproduced through the Birch equation using the parameters 69 GPa and 2.6 as reported (Léger, 1993).

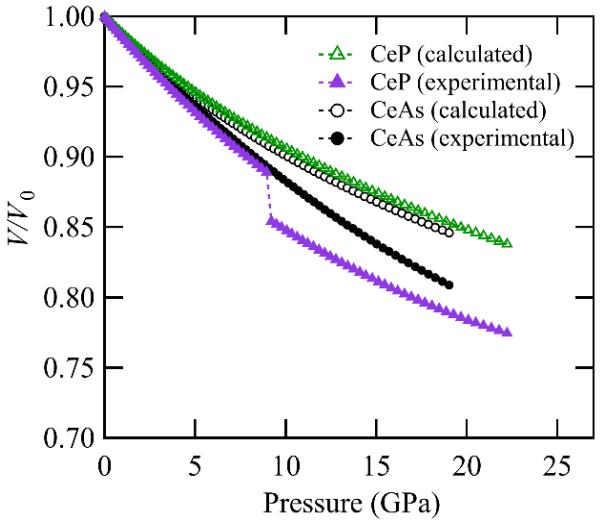


**Fig. 3. Reproduced (experimental) relative volume versus pressure curves of CeP and CeAs (filled triangles and filled circles) and their estimated curves (open triangles and open circles) for the existence of stable $Ce^{3+}$ ions in the materials.**

### Relative volume of CeSb and CeBi

The variation of reproduced experimental relative volume with pressure for CeSb and CeBi is shown in Fig. 4. For CeSb, the experimental data has exactly been reproduced through the Birch equation by using the parameters 71.5 GPa and 2.5 as reported by Léger (1993).

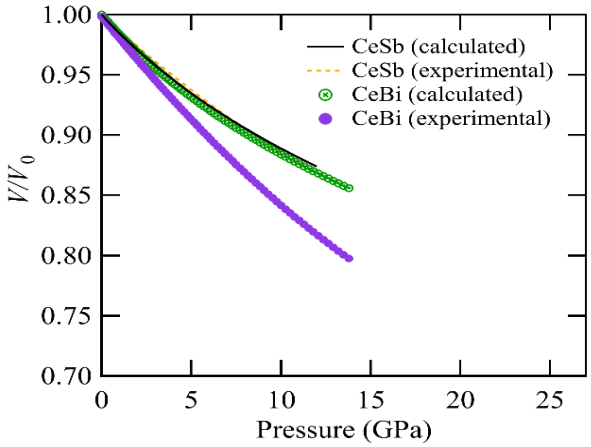


**Fig. 4. Reproduced (experimental) relative volume versus pressure curves of CeSb and CeBi (dashed line and filled circles) and the analogous calculated curves (solid line and crossed circles) for $Ce^{3+}$ ions in the materials.**

In the case of CeBi, the reproduced data using the parameters $B_0 = 50$ GPa and $B^* = 2.1$ in Eq. (1) precisely corresponds with the experimental data of Léger (1993). The parameters $B_0$ and $B^*$ used to reproduce the experimental relative volume with pressure for the compounds mentioned earlier are listed in Table 1, along with their lattice parameters at ambient pressure.

## Theoretical relative volume for $Ce^{3+}$

The relative volumes as a function of the pressure of the compounds CeO, CeS, and CeTe from the category of cerium monochalcogenides and the compounds CeP, CeAs, CeSb, and CeBi from cerium monopnictides have been calculated using the Birch equation for the stable trivalent cerium ions in them. The values of the parameters, the bulk modulus $B_0$ and its first pressure derivative $B^*$, as mentioned in Eq. (1), are required to know for performing the calculation described above.

The bulk modulus of any stable trivalent rare-earth compound can be obtained from the linear variations of the bulk moduli with the inverse of the volumes of unit cells of the compounds as reported (Léger, 1993). The parameters $B_0$ and $B^*$ for the calculation of relative volumes with stable $Ce^{3+}$ ions in the mentioned compounds are also listed in Table 1. Values of $B_0$ and $B^*$, listed in Table 1 against the mentioned pressure range were used in the calculation

**Table 1. Lattice parameter ($a$), bulk modulus ($B_0$) and its first pressure derivative ($B^*$) of some selected compounds**

| Compound | $a$ (Å) | For the reproduction of experimental relative volume | | | For the calculation of relative volume with stable $Ce^{3+}$ ions in the compound | | |
|---|---|---|---|---|---|---|---|
| | | Pressure Range (GPa) | $B_0$ (GPa) | $B^*$ | Pressure Range (GPa) | $B_0$ (GPa) | $B^*$ |
| CeO | 5.089[a] | 0 - 3 | 30 | 5 | All | 125.0[a] | 5.4[a] |
| | | 3 - 23.6 | 1.58 | 679 | | | |
| CeS | 5.766[a] | All | 83.5 | 2.2 | All | 85.0[a] | 5.4 |
| | 5.780[b] | | | | | 96.0[b] | 4.70[b] |
| | 5.730[c] | | | | | 87.0[c] | 4.83[c] |
| CeTe | 6.360[a] | All | 58 | 8 | All | 66.0[a] | 5.15 |
| | 6.360[c] | | | | | 58.2[c] | 5.15[c] |
| CeP | 5.920[a] | 0 - 9 | 64 | 3 | All | 78.0[a] | 5.4[a] |
| | | 9 - 22 | 30 | 9.2 | | | |
| CeAs | 6.080[a] | All | 69 | 2.6 | All | 72.4[a] | 5.4 |
| CeSb | 6.420[a] | All | 71.5 | 2.5 | All | 61.8[a] | 5.4 |
| CeBi | 6.550[a] | All | 50 | 2.1 | All | 58.2[a] | 5.4 |

[a]Reference (Léger, 1993)

[b]Reference (Svane et al., 1999)

[c]Reference (Bouhemadou et al., 2005)

The calculated relative volumes of CeO, CeS, and CeTe for stable $Ce^{3+}$ ions in them are shown in Fig.1 and Fig. 2 by open circles, open diamonds, and solid lines, respectively. Fig. 3 and Fig. 4 show the calculated relative volumes for CeP, CeAs, CeSb and CeBi by open triangles, open circles, solid lines, and crossed circles, respectively.

## Calculation of valence change

The valence of Ce in Ce$X$ ($X$ = O, S, Te, P, As, Sb, and Bi) is assumed to vary linearly with the lattice parameter, $a$ (Léger, 1993). According to this idea for a change in lattice parameter of $\{a^{3+}_{\text{calc}}(P_0) - a^{4+}_{\text{calc}}(P_0)\}$, the increase in valence is 1, where $a^{3+}_{\text{calc}}(P_0)$ and $a^{4+}_{\text{calc}}(P_0)$ are the estimated lattice constants at normal pressure with cerium valence 3+ and 4+, respectively. So, the increase in valence, $x$ ($x < 1$) above 3+ can be estimated by the equation,

$$x = \frac{\{a^{3+}_{\text{calc}}(P) - a^{3+x}_{exp}(P)\}}{\{a^{3+}_{\text{calc}}(P_0) - a^{4+}_{\text{calc}}(P_0)\}} \quad (2)$$

where $a^{3+}_{\text{calc}}(P)$ and $a^{3+x}_{exp}$ represent the calculated and experimentally observed lattice constants with cerium valence 3+ and 3+$x$, respectively.

Cerium monochalcogenides and monopnictides have *fcc* crystal structures, and each cerium ion has a coordination of six. In our calculation, the radii of $Ce^{3+}$ and $Ce^{4+}$ ions at ambient pressure are used as $r^{3+}_{\text{Ce}}(P_0)$=1.034Å, and $r^{4+}_{\text{Ce}}(P_0)$=0.85 Å, respectively, as reported (Léger, 1993). The calculated lattice parameter for a compound with $Ce^{3+}$ ions at ambient pressure $P_0$ can be expressed as $a^{3+}_{\text{calc}}(P_0) = 2r^{3+}_{\text{Ce}}(P_0) + 2r_X(P_0)$, where $X$ = O, S, Te, P, As, Sb, Bi and $r_X(P_0)$ is the ionic radius of the concern anion. So, the change in lattice parameter between its two ionic states ($Ce^{3+}$ and $Ce^{4+}$) of cerium in a compound is written as

$$a^{3+}_{\text{calc}}(P_0) - a^{4+}_{\text{calc}}(P_0) = 2\{r^{3+}_{\text{Ce}}(P_0) - r^{4+}_{\text{Ce}}(P_0)\} = 2(1.034 - 0.85)\ \text{Å} = 0.368\ \text{Å}$$

Thus, Eq. (2) becomes,

$$x = \frac{\{a^{3+}_{\text{calc}}(P) - a^{3+x}_{exp}\}}{0.368} \quad (3)$$

$$\text{Or, } x = \frac{a^{3+}_{\text{calc}}(P_0)\left\{\frac{a^{3+}_{\text{calc}}(P)}{a^{3+}_{\text{calc}}(P_0)} - \frac{a^{3+x}_{exp}}{a^{3+}_{\text{calc}}(P_0)}\right\}}{0.368}, \quad (4)$$

$$\text{Or, } x = \frac{a^{3+}_{\text{calc}}(P_0)\left[\left\{\frac{V_{\text{calc}}(P)}{V(P_0)}\right\}^{\frac{1}{3}} - \left\{\frac{V_{\text{exp}}(P)}{V(P_0)}\right\}^{1/3}\right]}{0.368}, \quad (5)$$

where $V(P_0) = \left\{a^{3+}_{\text{calc}}(P_0)\right\}^3$, $V_{\text{calc}}(P) = \left\{a^{3+}_{\text{calc}}(P)\right\}^3$ and $V_{\text{exp}}(P) = \left\{a^{3+x}_{\text{exp}}(P)\right\}^3$ are the estimated volume of the unit cell at ambient pressure with $Ce^{3+}$ ion, the estimated volume at pressure $P$ with $Ce^{3+}$ ion and the experimentally found volume at pressure $P$ with $Ce^{3+x}$ ion, respectively.

The Ce valence at any pressure $P$ is calculated by the relation

$$\text{Valence} = 3 + \frac{a^{3+}_{\text{calc}}(P_0)\left[\left\{\frac{V_{\text{calc}}(P)}{V(P_0)}\right\}^{1/3} - \left\{\frac{V_{\text{exp}}(P)}{V(P_0)}\right\}^{1/3}\right]}{0.368} \quad (6)$$

## Results and Discussion

The calculated valences, using Eq. (6), of Ce in CeO, CeS, and CeTe with pressure are shown in Fig. 5. To calculate the valence number of the Ce in CeO, the experimental relative volume $V_{\text{exp}}(P)/V(P_0)$ has been reproduced by the Birch Eq. (1) as shown in Fig. 1. As the reproduction is incompatible with experimental data as reported by Léger (1993) in the pressure range about 3 to 6 GPa, the valence has been calculated up to 23.6 GPa except for the mention pressure range. As represented by solid triangles, the valence of Ce rises spectacularly with the increase of pressure up to 3 GPa, where the valence is 3.26+. As the pressure increases above 6 GPa, the valence decreases slowly with the pressure and reaches 3.16+ at 23.6 GPa. For CeS, the valence remains almost constant with pressure up to about 4 GPa, but above this pressure, the valence increases almost steadily and reaches 3.27+ at about 26 GPa. Starting from 3.0+ at ambient pressure, the valence of cerium in CeTe exhibits a slightly upward trend with increasing pressure but falls again to 3.0+ at around 8 GPa.

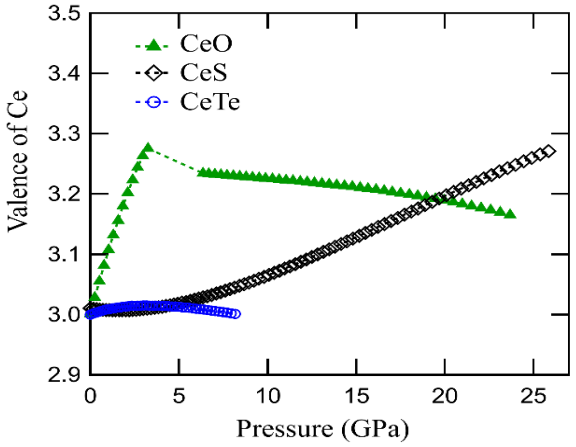


**Fig. 5. Calculated valence of Ce as a function of pressure in CeO, CeS, and CeTe.**

Fig. 6 shows the calculated valences of Ce in CeP, CeAs, CeSb, and CeBi as a function of pressure. The valence of Ce in CeP grows almost consistently with the increase of pressure until it reaches 3.14+ at nearly 9 GPa. After that, the valence rises dramatically to 3.33+ at about 9.2 GPa because of the isostructural valence-phase transition. From 9.2 to 22.2 GPa, the valence of Ce exhibits no significant change with pressure. For CeAs, the valence of Ce increases non-linearly with pressure from 0 to about 5 GPa. Later, the valence increases almost linearly with further increasing pressure, and at pressure 19 GPa the valence is 3.22+. The valence of Ce, in the case of CeSb, decreases slightly and then grows with pressure. At a pressure of about 12 GPa, the valence is 3.02+. For CeBi, the valence of Ce increases gradually with pressure, and the valence is 3.39+ at around 14 GPa.

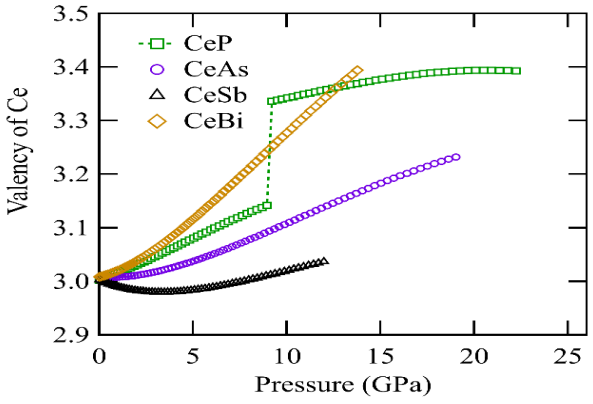


**Fig. 6. Calculated valence of Ce as a function of pressure in CeP, CeAs, CeSb, and CeBi.**

Our analysis reveals that the distinction between the experimental and calculated pressure-volume curves, as presented in section 2, is due to the valence change of Ce in cerium compounds with pressure. The valence of Ce in any group of compounds does not always increase with the increase of pressure but is also seen to decrease with pressure, as in the cases of CeO, CeTe, and CeSb. The overall trend of the valence changes of Ce in CeAs and CeBi is similar, but the rate of enhancement of valence with pressure in CeBi is much higher than that of CeAs. For the Ce-based compounds Ce*X*, either cerium monochalcogenides or cerium monopnictides, the valence change of Ce does not follow any unique rule, but depends on the constituted *X* ion, i.e., the particular environment around the Ce ion.

In the case of CeO/CeS/CeTe, the average electron transfer from $4f^1$ orbital of Ce to $3s^1/4s^1/6s^1$ orbital of O/S/Te with a certain increase of pressure gives the valence change of Ce at that pressure. In the case of CeP/CeAs/CeSb/CeSb, the average transfer of an electron from $4f^1$ orbital of Ce to $4s^0/5s^0/6s^0/7s^0$ orbital of P/As/Sb/Bi with a certain increase of pressure gives the valence change of Ce at that pressure. That means the valence change of Ce at any high pressure depends on the amount of the mixing of $4f^1$ orbital of the cation $Ce^{3+}$ with the s orbital of the anion at that pressure.

Our finding of the valence change of Ce in cerium monochalcogenides and monopnictides may be useful to the theoreticians to interpret the various properties of these compounds.

## Conclusions

The valence change of Ce in Ce*X* (*X*=O, S, Te, P, As, Sb, and Bi) with pressure has been successfully investigated by employing the Birch equation of state. For both CeP and CeBi, the maximum valence change is 0.39+ for the pressure change of 22.2 GPa and 13.8 GPa, respectively. It is found that the valence of Ce in the majority of the compounds increases with the increase of pressure. However, cerium-based compounds cannot ensure a proportionate increase or decrease of cerium valence with pressure. The valence change behavior of cerium with pressure in cerium-based compounds is a unique feature that depends only on the environment around the Ce ion.

## Authors' Contributions

Conceptualization: A.H. and M.T.H.; calculation: A.H., M.T.H. and M.S.I.; analysis: A.H., M.T.H. and M.S.I.; original draft preparation: M.S.I. and A.H.; reviewing and editing: A.H., M.T.H. and M.S.I; funding acquisition: A.H. All authors read and approved the final manuscript.

## Conflict of Interest

The authors declared that they have no conflict of interest.

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