Supplementary Materials for "Temperature-Dependent Elastic Properties of B_4C from First-Principles Calculations and Phonon Modeling"

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1. Thermal Properties from Phonon Calculations

Performing phonon calculations for each volume of B_4C yields phonon density of states and phonon dispersion curves, which can then be used to determine temperature-dependent thermal properties (Helmholtz free energy, entropy, and heat capacity at constant volume). Figure S1 illustrates these thermodynamic properties for B_4C at different volumes. It presents the variations of free energy, entropy, and heat capacity at constant volume (C_V) as functions of temperature. The trends of these thermodynamic properties arise from the anharmonic nature of atomic vibrations within the crystal lattice [1]. This anharmonicity becomes particularly significant at higher temperatures, where it influences the thermal expansion coefficient and phase transition [1]. The free energy (Figure S1(a)) decreases with increasing temperature due to the greater availability of phonon modes (vibrational states) at higher temperatures [2]. As the temperature rises, more vibrational modes become thermally accessible [2, 3], increasing the system's entropy (Figure S1(b)). C_V also increases with temperature due to the additional vibrational modes becoming accessible. At higher temperatures, the system gains more vibrational degrees of freedom, and the phonon modes contribute more significantly to the system's internal energy. Consequently, the system's internal energy follows classical behavior, as described by the Dulong-Petit law. This classical limit is illustrated in Figure S1(c).

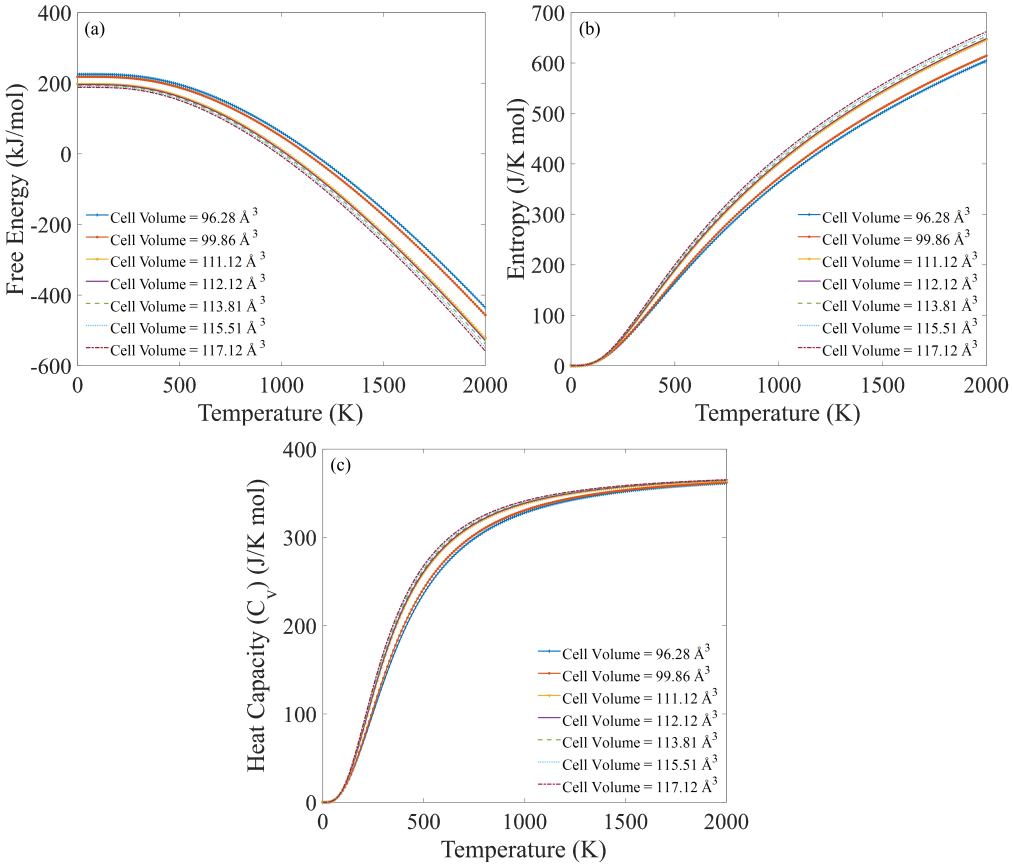

Figure S1 – Temperature-dependent thermodynamic properties of B4C for different volumes. (a) Helmholtz free energy, (b) entropy, and (c) heat capacity at constant volume.

References

[1] E. T. Ritz, S. J. Li, N. A. Benedek, Thermal expansion in insulating solids from first principles, Journal of Applied Physics 126 (17) (2019).

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[3] A. J. McGaughey, A. Jain, H.-Y. Kim, B. Fu, Phonon properties and thermal conductivity from first principles, lattice dynamics, and the boltzmann transport equation, Journal of Applied Physics 125 (1) (2019).