DETERMINATION OF HEAT CAPACITY OF SOME SEMICONDUCTOR COMPOUNDS USING A DELAUNAY'S INTEGRAL

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Abstract

Using a numerical Delaunay's method, theoretical values of molar heat capacity C_p were calculated for some semiconductors (GeS, GeSe, GeTe). The obtained results have been compared with the n-dimensional Debye functions and with the corresponding experimental results in the 220-250 K temperature range. The estimated results are in good agreement with both the n-dimensional Debye functions and the experimental results over the entire temperature range. This confirms that the numerical Delaunay's method is able to represent the molar heat capacity C_p of semiconductor compounds.

Keywords: Semiconductor Compounds; Molar Heat Capacity; Numerical Delaunay's Method; Debye's Temperature.

Introduction

The chalcogenides of group (IV-VI) (GeS, GeSe, GeTe, etc) have aroused considerable recent attention due to their extraordinary properties such as inharmonic effects (Delaire et al., 2011), weak lattice thermal conductivity (Pei et al., 2011; Wang et al., 2014), unprecedented thermoelectric performance (Wang et al., 2011; Pei et al., 2012; Chen et al., 2014; Wu et al., 2014), complex band structure (Cagnoni et al., 2018), chemical ultrahigh power factor (Zhao et al., 2016), and chemical stability, and a lesser degree of toxicity (Qin et al., 2016). Some of these compounds display ferroelectricity, paraelectricity, and superconductivity (Zhang et al., 2019; Ronneberger et al., 2020; Gui et al., 2020). In addition, most IV–VI compounds have a band gap size (Cohen and Chelikowsky, 1988) that makes them suitable for devices such as infrared detectors and lasers, nanoelectronics, and thermoelectrics (Ding et al., 2015; Patel et al., 2017).

The mathematician Delaney was the first to use numerical methods to solve the Debye integral relationship. This method is completely generic and free of any restrictions on its application to analyze the thermo-physical properties of binary semiconductor (Delaunay, 1956; Aldeeb, 2017).

The heat capacity varies from one substance to another; this difference is due to the extent of the atoms of the substance and their ability to conduct heat (Eugène, 2009). Therefore, determining the heat capacity of crystals of semiconducting materials is one of the important topics of solid physics, and determining heat capacity of any semiconductor material gives an insight into its vibrational properties (Nernst et al., 1911; Kittel, 1956). Furthermore, it will be useful to know many thermodynamic properties of these materials such as entropy, enthalpy, and others.

These reasons prompted many researchers to study heat capacity in two ways: the experimental (traditional) method and the theoretical method. Bevolo et al. (1976) experimentally determined the heat capacity of GeTe, SnTe and PbTe compounds in the 0.9 K to 60 K temperature range by the heat-pulse method. In addition, Wiedemeier et al. (1978) measured the heat capacities of the GeS, GeTe and GeSe compounds by using a computer interfaced differential scanning calorimeter.

Various efficient theoretical methods were proposed to determine the molar heat capacity C_p of semiconductors. Some of them relied on n-dimension Debye functions, some on the Lambert-Boltzmann distribution, others on the Einstein's model, some of them combined Debye's and Einstein's model, and others used a numerical Delaunay's method derived from Debye's model. Here, the most important studies that dealt with these compounds will be presented. Eser et al. (2011) estimated some theoretical values of heat capacity C_p for some semiconductors using n-dimensional Debye functions. Boussaid et al. (2019) determined the heat capacity of some semiconductor compounds by the Lambert W function. Algeidi and Aldeeb (2021) calculated the molar heat capacity C_p of the SnSe semiconductor compound using a numerical Delaunay's method. All these studies reached satisfactory results, and the semiconductor compounds under study have been theoretically studied using n-dimensional Debye functions.

The aim of this study is to calculate the heat capacity C_p of the semiconductor compounds, namely Germanium sulfide (GeS), Germanium selenide (GeSe) and Germanium telluride (GeTe). Temperature range from will be 220 K to 250 K, based on Delaunay's method Chromatography. The results will be compared with the experimental data (Wiedemeier et al., 1978) and with theoretical values of C_p calculated by n-dimensional Debye functions (Eser et al., 2011).

Crystal Structure and Properties

The germanium sulfide, germanium selenide and germanium telluride compounds are binary semiconductors which follow the orthorhombic crystal system. They all convert to the cubic crystal system (NaCl structure) (Schubert,1964), but the transformation process (transitions phase) varies from one compound to another depending on the temperature. Accordingly, phase transitions from the rhombohedral to the NaCl structure was observed for GeTe compound at about 670 K (Zhukova and Zaslavski, 1967). While the orthorhombic compounds GeS and GeSe behave differently, GeSe transforms at about 924 K to the NaCl structure in one step. However, this compound reveals certain anomalies of thermal expansion in the 670 – 770 K temperature range (Wiedemeier and Siemers, 1975). GeS compound displays similar anomalies of the thermal expansion in the temperature range of about 500 – 780 K, and melts at 931 K before the possible transition to the NaCl structure (Wiedemeier and Siemers, 1977). These compounds (GeS, GeSe and GeTe) have melting temperature of $(931\ \mathrm{K}, 948\ \mathrm{K}, 996\ \mathrm{K})$ respectively, and they have Debye's temperature of $(360\ \mathrm{K}, 280\ \mathrm{K}, 180\ \mathrm{K})$ respectively (Wiedemeier et al., 1978).

Expression for Molar Heat Capacity (C_p)

Debye's model of heat capacity at constant volume (C_v) is given by (Ashcroft and Mermin, 1976; Moustafa, 2007):

$$c_{\mathrm{v}} = \left(\frac{\partial u}{\partial T}\right)_{\mathrm{v}} = \frac{9ZN_A\hbar^2}{\omega_D^3 k_B T^2}\int_0^{\omega_D} \frac{\omega^4 e^{\frac{\hbar\omega}{k_B T}} d\omega}{\left(e^{\frac{\hbar\omega}{k_B T}} - 1\right)^2}, \qquad (1)$$

Where Z is the number of atoms of one primitive cell (number of atoms in a molecule of the material), N_A is the Avogadro number, T is the absolute temperature, ω_D is the Debye's greatest angular frequency, k_B is the Boltzmann constant, ω is the angular frequency. And $\hbar = \mathrm{h}/2\pi$, where h is Planck's constant (Ashcroft and Mermin, 1976; Moustafa, 2007). This integral equation is difficult to integrate mathematically, but Delaunay succeeded to solve it numerically. Based on mathematical substitutions and $\omega = P\omega_D$, Delaunay reduces the equation (1) to the following form (Delaunay, 1956):

$$c_{\mathrm{v}} = \frac{9ZN_A\hbar^2{\omega_D}^2}{k_B T^2}\int_0^1 \frac{P^4 e^{(\hbar\omega_D/k_B T)P} dP}{(e^{(\hbar\omega_D/k_B T)P} - 1)^2}, \qquad (2)$$

Since the $k_B\theta_D = \hbar\omega_D$,$\theta_D$ is the Debye's temperature, and by putting $x = T/\theta_D$&$y = C_{\mathrm{v}}/3ZR$,R is the gas constant, we get (Delaunay, 1956):

$$y = \frac{3}{x^2}\int_0^1 \frac{P^4 e^{P/x} dP}{(e^{P/x} - 1)^2}, \qquad (3)$$

Using integration, we get:

$$y = \frac{12}{x}\int_0^1 \frac{P^3 dP}{e^{P/x} - 1} - \frac{3}{x}\left(\frac{1}{e^{1/x} - 1}\right), \qquad (4)$$

From equation (4), we get molar heat capacity C_v, and to determine the molar heat capacity C_p from C_v, one may use the Nernst-Lindemann equation (Nernst et al., 1911; Rabe and Joannopoulos, 1985), the resultant equation is:

$$C_p(T) - C_v(T) = A_0\left[C_p(T)\right]^2 \frac{T}{T_m}, \quad (5)$$

Which:

$$C_p(T) = \frac{T_m}{2A_0T}\left[1 - \left[1 - 4A_0\frac{T}{T_m}C_v(T)\right]^{\frac{1}{2}}\right], \quad (6)$$

Where T_m is the melting temperature, $A_o = 5.1 * 10^{-3}\ [J^{-1}.mol \cdot K]$. The value A_o is relatively small magnitude, then the square root in equation (6) can be expanded conveniently into a Taylor series expansion, thus equation (6) can be written as follows (Pässler, 2007; Schick, 2013):

$$C_p(T) = C_v(T) + A_0[C_v(T)]^2\frac{T}{T_m}, \quad (7)$$

And equation (7) can be written as:

$$C_p(T) = 3ZR\left\{\frac{12}{x}\int_0^1 \frac{P^3dP}{e^{P/x} - 1} - \frac{3}{x}\left(\frac{1}{e^{1/x} - 1}\right)\right\} + A_0\left[3ZR\left\{\frac{12}{x}\int_0^1 \frac{P^3dP}{e^{P/x} - 1} - \frac{3}{x}\left(\frac{1}{e^{1/x} - 1}\right)\right\}\right]^2\frac{T}{T_m}, \quad (8)$$

Result and Discussion

Using the MatLab software, Delaunay's numerical method was applied to calculate the theoretical values of molar heat capacity at constant pressure (C_p) for some binary semiconductor compounds such as germanium sulfide (GeS), germanium selenide (GeSe) and germanium telluride (GeTe) in the 220 - 250 K temperature range, depending on the Debye's temperature and the melting temperature of each compound mentioned earlier. The obtained results for GeTe, GeSe and GeS compounds were compared with their experimental measurements (Wiedemeier et al., 1978), which are shown in Tables (1), (2) and (3), respectively. It is evident from these tables that the theoretical results for the molar heat capacity C_p of semiconductor compounds are in good agreement with the experimental data (Wiedemeier et al., 1978) for all values of the temperature for each compound with a discrepancy of 1.84% for semiconductor compound GeS, 0.4% for GeSe compound, and finally 0.3% for GeTe compound. They were all at 250 K, as shown in Figures (1), (2) and (3), respectively. In comparison with the data available in the literature showed in Tables (4), (5) and (6), the results are in excellent agreement with the calculated values by Eseretal. (2011) using n-dimensional Debye functions for the semiconductor compounds $(\mathrm{GeTe}, \mathrm{Ge}S\mathrm{e}, \mathrm{GeS})$ within that range at the same melting temperature and Debye's temperature for each compound, as shown in Figures (4), (5) and (6), respectively. Moreover, this result is quite similar to that, which the researcher conducted previously to calculate the molar heat capacity C_{pc} of tin selenide (SnSe) in the temperature range 20 - 500 K. In that study, it had been almost reached excellent agreement between the theoretical calculations based on this numerical method and experimental data, n-dimension Debye function, and Lambert-Boltzmann distribution, for the range of these temperatures (Algeidi and Aldeeb, 2021).

Figures (7), (8) and (9) show respectively the comparison of the theoretical values of molar heat capacity at constant pressure obtained from the numerical Delaunay's method with experimental data (Wiedemeier et al., 1978) and with the results of Eser et al. (2011) for semiconductor compounds $\mathrm{GeTe}, \mathrm{Ge}S\mathrm{e}$ and GeS, respectively. These figures show the great congruence between them. This shows that the numerical Delaunay method derived from Debye's model is very effective in finding theoretical values for the molar heat capacity at constant pressure (C_p) for binary semiconductors compounds.

Table (1): The Comparative Results of Molar Heat Capacity $C_p(T)$ of GeS Compound at $\theta_D = 360\ K$ and $T_m = 931\ K$ with Experimental Data.

T [K]	Our Calculated C_p [J/(mol.K)]	(Wiedemeier *et al.*, 1978) C_p [J/(mol.K)]	Relative Error
220.00	46.1092	45.6100	0.01094
230.00	46.7424	46.1100	0.01372
240.00	47.3226	46.5700	0.01616
250.00	47.8569	46.9900	0.01845

Table (2): The Comparative Results of Molar Heat Capacity $C_p(T)$ of GeSe Compound at $\theta_D = 280\ K$ and $T_m = 948\ K$ with Experimental Data.

T [K]	Our Calculated C_p [J/(mol.K)]	(Wiedemeier *et al.*, 1978) C_p [J/(mol.K)]	Relative Error
220.00	48.5809	48.4700	0.00229
230.00	49.0385	48.8900	0.00304
240.00	49.4599	49.2700	0.00385
250.00	49.8503	49.6200	0.00464

Table (3): The Comparative Results of Molar Heat Capacity $C_p(T)$ of GeTe Compound at $\theta_D = 180$ K and $T_m = 996$ K with Experimental Data.

T [K]	Our Calculated C_p [J/(mol.K)]	(Wiedemeier *et al.*, 1978) C_p [J/(mol.K)]	Relative error
220.00	50.8794	50.8200	0.00117
230.00	51.1498	51.0400	0.00215
240.00	51.4036	51.2600	0.00280
250.00	51.6431	51.4800	0.00317

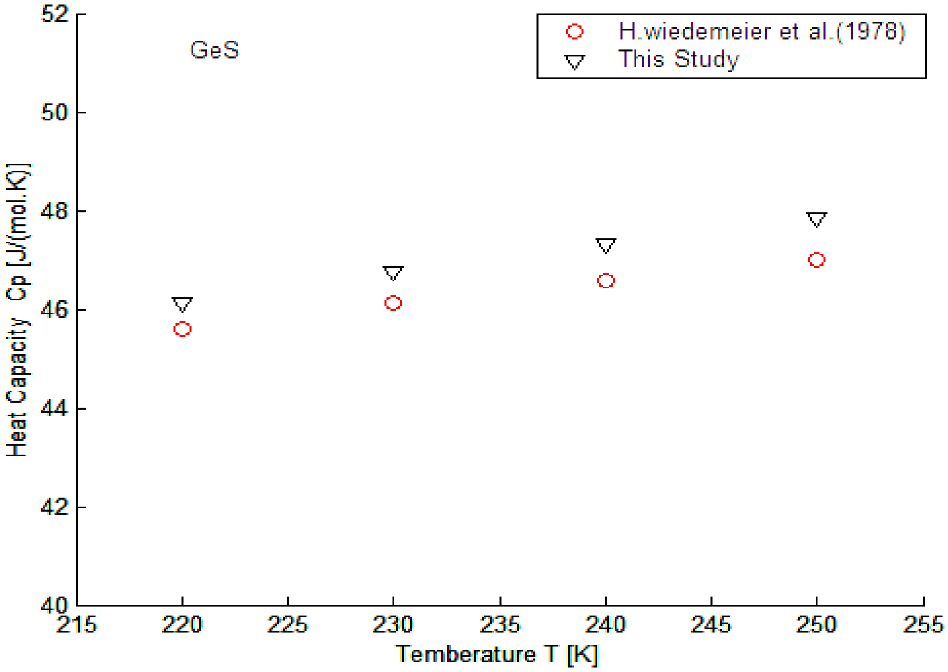

Figure (1): Comparison between the Experimental Data and the Theoretical Values of the Molar Heat Capacity C_p Resulting from Delaunay's Method for GeS Compound at $\theta_D = 360$ K and $T_m = 931$ K.

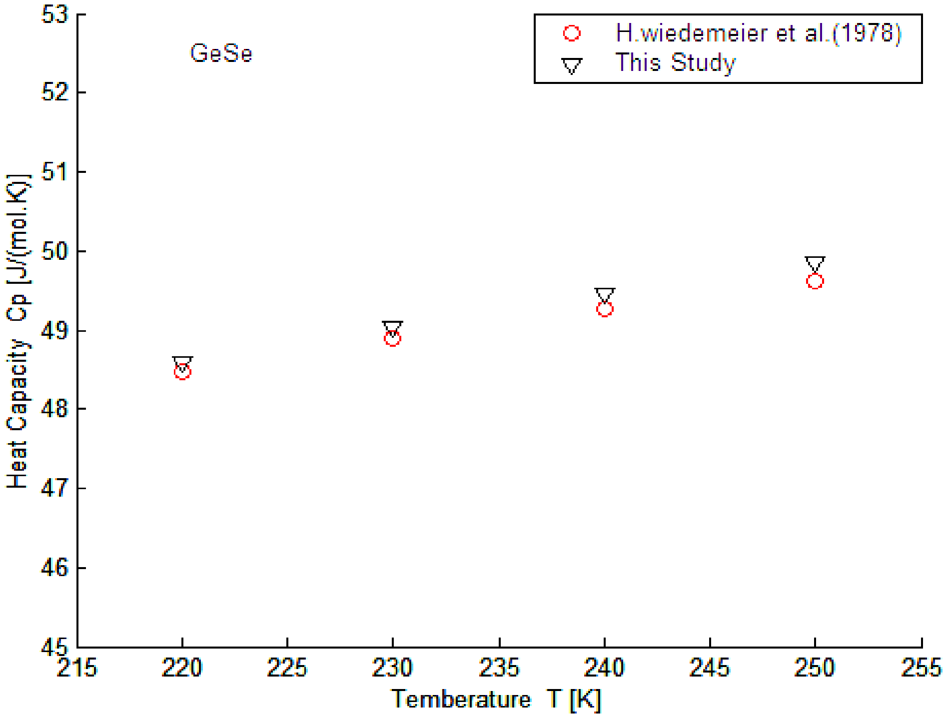

Figure (2): Comparison between the Experimental Measurements and the Theoretical Values of the Molar Heat Capacity C_p Resulting from Delaunay's Method for GeSe Compound at $\theta_D = 280$ K and $T_m = 948$ K.

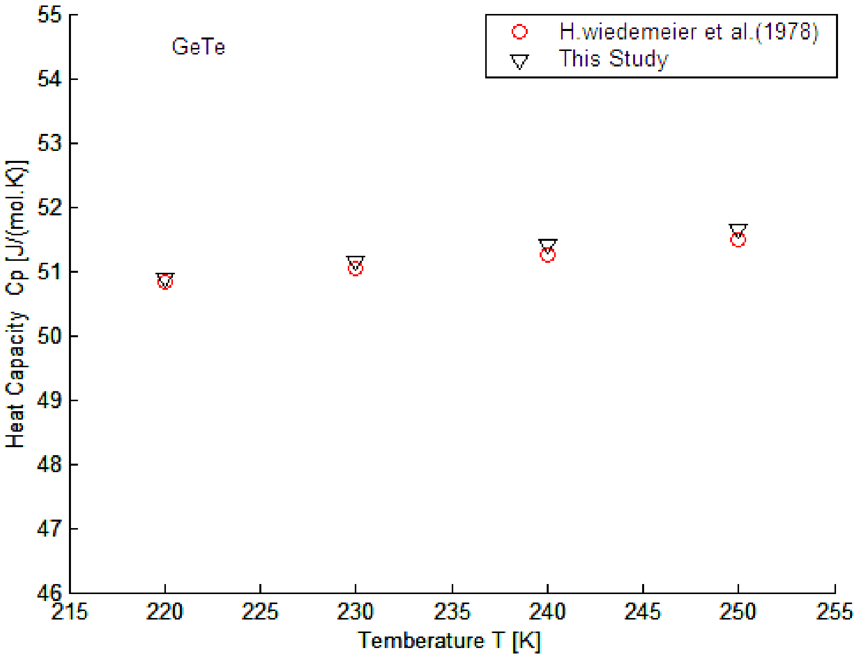

Figure (3): Comparison between the Experimental Measurements and the Theoretical Values of the Molar Heat Capacity C_p Calculated from Delaunay's Method for GeTe Compound at $\theta_D = 180$ K and $T_m = 996$ K.

Table (4): Comparison of the Theoretical Values of Molar Heat Capacity C_p Calculated by Delaunay's Solution for GeS with the Theoretical Values Obtained by Eser et al.

T [K]	Our Calculated C_p [J/(mol.K)]	(Eser *et al.*, 2011) C_p [J/(mol.K)]	Relative error
220.00	46.1092	46.1090	0.000004
230.00	46.7424	46.7420	0.000009
240.00	47.3226	47.3220	0.000013
250.00	47.8569	47.8570	0.000002

Table (5): Comparison between the Molar Heat Capacity C_p Values Calculated by Delaunay's Law for GeSe with the Theoretical Values for Eser et al.

T [K]	Our Calculated C_p [J/(mol.K)]	(Eser *et al.*, 2011) C_p [J/(mol.K)]	Relative error
220.00	48.5809	48.5810	0.000002
230.00	49.0385	49.0380	0.00001
240.00	49.4599	49.4600	0.000002
250.00	49.8503	49.8500	0.000006

Table (6): Comparison of the Theoretical Values of Molar Heat Capacity C_p Calculated by Delaunay's Law for GeTe with the Theoretical Values Result by Eser et al.

T [K]	Our Calculated C_p [J/(mol.K)]	(Eser *et al.*, 2011) C_p [J/(mol.K)]	Relative error
220.00	50.8794	50.8790	0.000008
230.00	51.1498	51.1490	0.000016
240.00	51.4036	51.4030	0.000012
250.00	51.6431	51.6430	0.000002

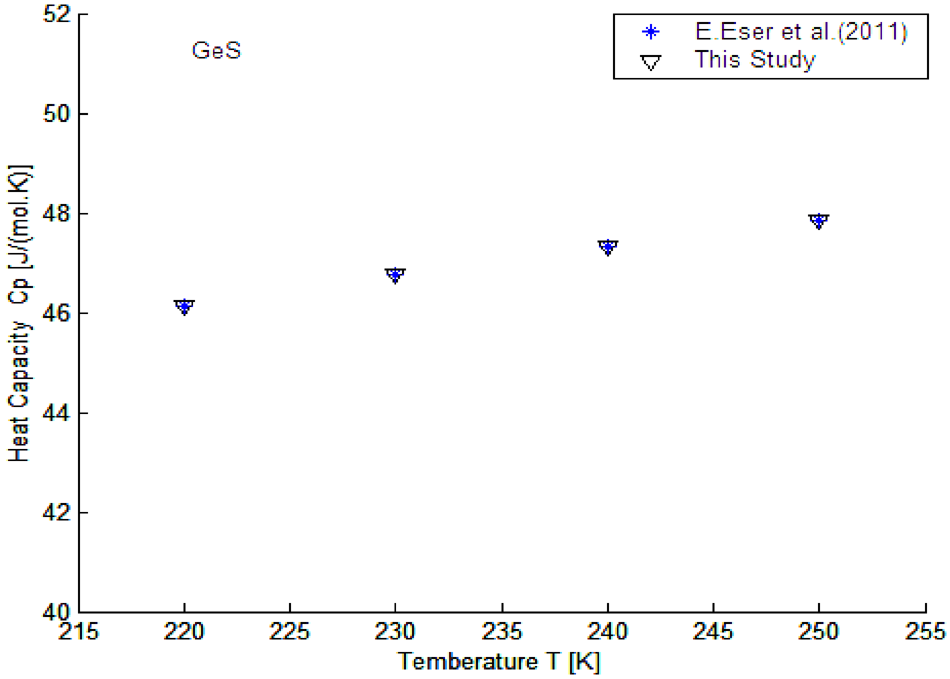

Figure (4): Comparison between the Theoretical Points According to Delaunay's Method and Theoretical Points of n-Dimension Debye Functions for the Molar Heat Capacity Cp for GeS Compound.

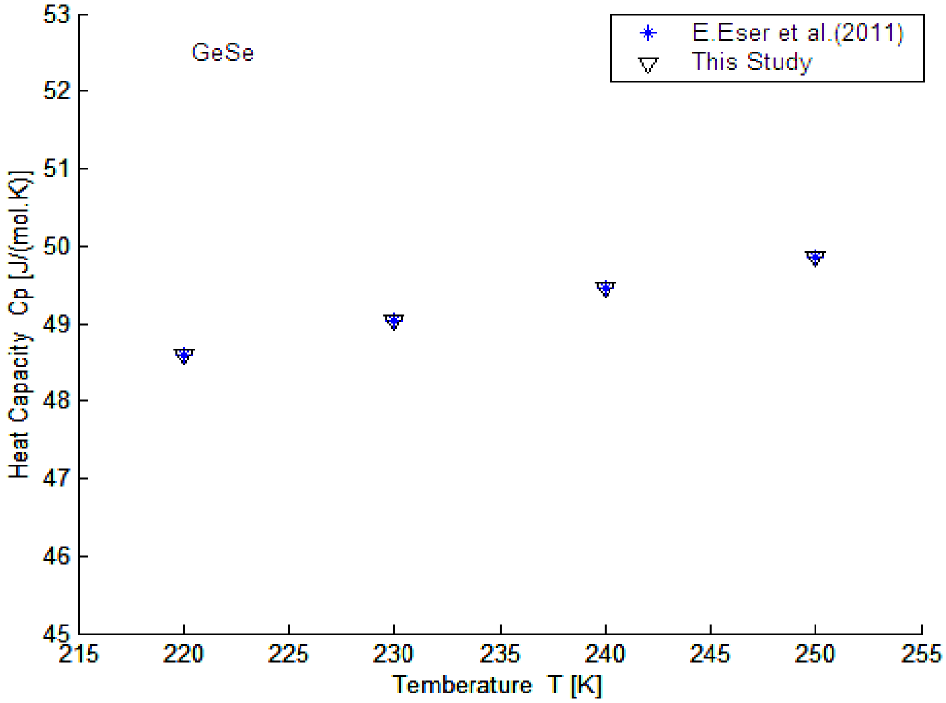

Figure (5): Comparison between the Theoretical Values Results from Numerical Delaunay's Method and Theoretical Points of n-Dimension Debye Functions of the Molar Heat Capacity C_p for GeSe Compound.

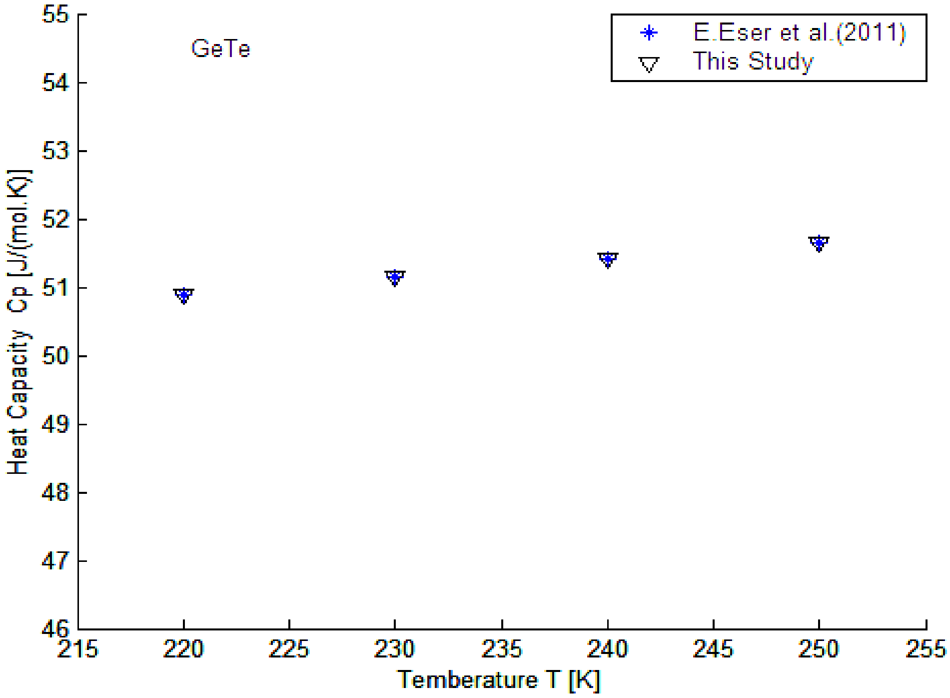

Figure (6): Comparison the Theoretical Values Result from Delaunay's Method and Theoretical Values of n-Dimension Debye Functions of the Molar Heat Capacity C_p for GeTe Compound.

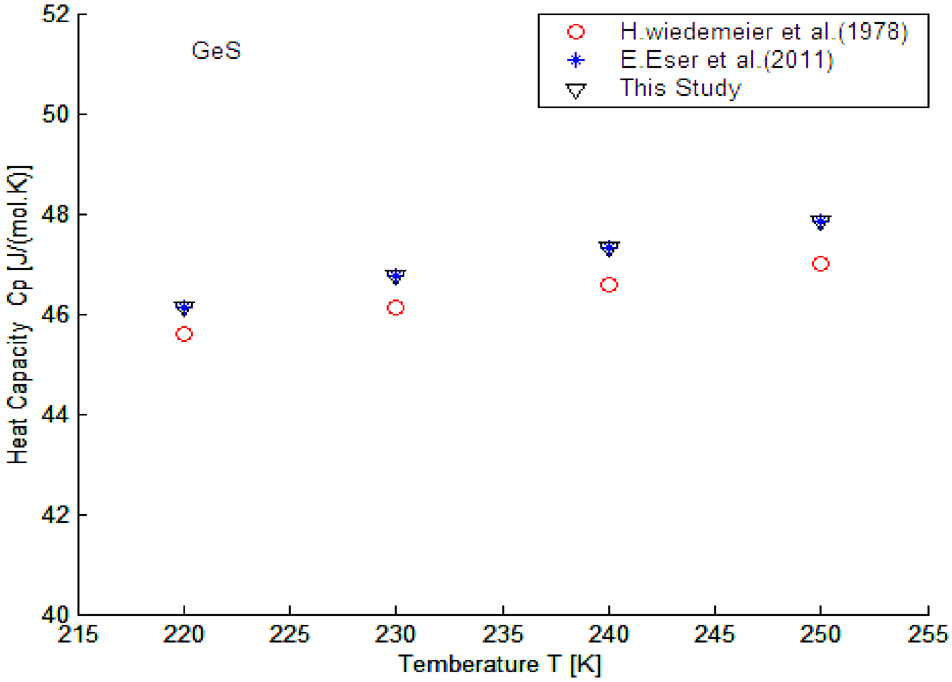

Figure (7): Comparison of the Theoretical Values of Molar Heat Capacity C_p Obtained from Delaunay's Method with Experimental Data and with Theoretical Values of n-Dimension Debye Functions for GeS Compound.

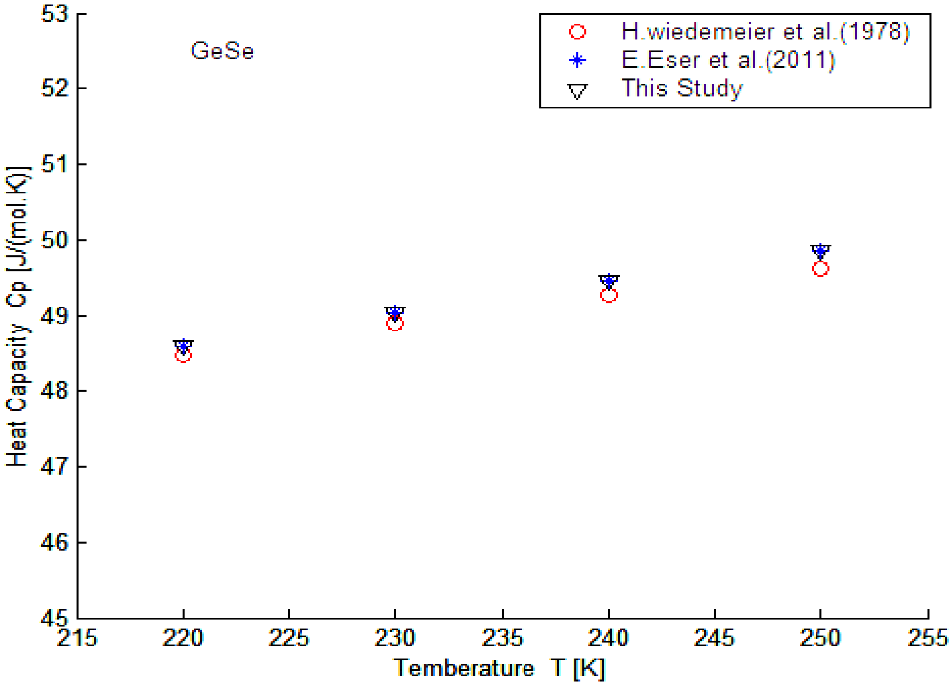

Figure (8): Comparison of the Theoretical Values of Molar Heat Capacity C_p According to the Delaunay's Integration with Experimental Measurements and with Theoretical Values of n-Dimension Debye Functions for GeSe Compound.

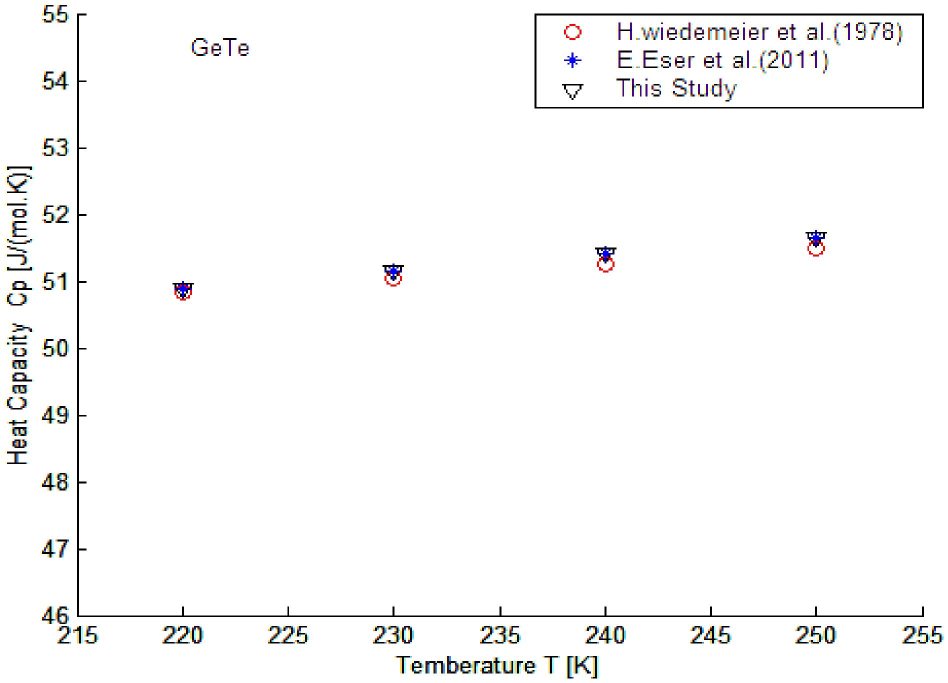

Figure (9): Comparison of the Theoretical Values of Molar Heat Capacity C_p Obtained from the Delaunay's Integration with Experimental Measurements and with Theoretical Values of n-Dimension Debye Functions for GeTe Compound.

Conclusion

The mathematical Delaunay's integration was used to determine the molar heat capacity C_p of binary semiconductor compounds (GeTe, GeSe, GeS) in the 220 – 250 K temperature range. The obtained results have been compared with experimental data, and theoretical values determined by Eser et al. using n-dimensional Debye functions for each compound. The estimated results are in good agreement with both the n-dimensional Debye functions and the experimental results over the entire temperature range. The numerical Delaunay's solution derived from the Debye's integration was successful in determining the molar heat capacity at constant pressure (C_p) of the binary semiconductor compounds. The researcher recommends using it to calculate of the molar heat capacities C_p of other semiconductor compounds and using it when studying the total internal energy and linear thermal expansion coefficients.

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