COMPARATIVE STUDY OF THEORETICAL AND EXPERIMENTAL HEAT CAPACITY CV FOR CADMIUM TELLURIDE

Basmah A. Aldeeb

Department of Physics, Abu-Isa Faculty of Education, University of Zawia, Libya
b.aldeeb@zu.edu.ly

Abstract

The aim of this study was to compare theoretical values with experimental measurements of molar heat capacity at a constant volume (C_v) in order to determine the best value for a Debye's temperature (θ_D) for cadmium telluride (CdTe). Debye's model was used to calculate the molar heat capacity C_v as a function of the absolute temperature for CdTe in the temperature ranges 7 K to 700 K. The experimental measurements were compared with the calculated values using several Debye's temperatures to determine the best congruence between the calculated and the experimental results of heat capacity C_v.

The results showed great agreement between the calculated theoretical values of C_v and the experimental data at medium and high temperatures. In general, the Debye's temperature of 144.67 K gave a best fit between the experimental and the theoretical results almost in the entire temperature range, where the sum of squares of deviations was $0.3824\ [\mathrm{J}/(\mathrm{mol}\cdot\mathrm{K})]^2$.

Keywords: CdTe; Experimental; Heat capacity; Debye's model; Debye-temperature.

Introduction

Among II–VI semiconductor compounds, Cadmium telluride (CdTe) is of special technological interest and a typical representative of the wide band-gap semiconductor material. Wide-band-gap semiconductors are of scientific and technological interest, and they have gained popularity as a key material for applications in high-performance optoelectronic and electronic devices (Granqvist, 2007; Hosono, 2007). CdTe emerged as a promising material for room-temperature X- and gamma-ray detectors (Szeles, 2004; Li et al., 2012). These applications include industrial monitoring, gauging and imaging, medical imaging, and transportation security and safety devices (Britt and Ferekides, 1993; Szeles, 2004; Li et al., 2012). Besides, it is used to make thin-film solar cells (Yoo et al., 2014), infrared optical materials for optical windows and lenses, infrared detection (Wald, 1977; Wu, 2004; Munshi, 2018) and electro-optic modulators (Valmik et al., 2021). The Debye's model of heat capacity is considered the most successful model for describing the graphic behavior of heat capacity at a constant volume (C_v) as a function of absolute temperature (T) of solid crystals. Its results concluded to an integrative relationship of C_v which is difficult to integrate mathematically. Using numerical methods, Delaney was able to solve the Debye's integral relationship. He made this relationship not dependent on a particular type of material and also free of any restrictions on its application to analyze the thermo-physical properties of binary semiconductor (Delaunay, 1956; Aldeeb, 2017; Algeidi and Aldeeb, 2021; Aldeeb, 2022).

Therefore, the Delaney numerical methods will be used to find the theoretical values of the heat capacity of CdTe, and to estimate the Debye's temperature. The Debye's temperature θ_D is the temperature of the crystal's highest normal mode of vibration (Debye, 1912; Hill, 1986) i. e. the highest temperature that can be achieved due to a single normal vibration (Hill, 1986). Determination of the value of Debye's temperature (θ_D) for any substance is one of the important topics of solid-state physics. The Debye's temperature is the key concept to correlate the elastic properties with thermodynamic properties, such as phonons, thermal expansion, thermal conductivity, heat capacity, and lattice enthalpy (Li and Wang, 2012; Tosto, 2016), also to correlate with optical and electric properties of solids (Tosto, 2016). The heat capacity is an important thermodynamic characteristic of matter. Therefore, reliable data on the heat capacity of CdTe (Pavlova, 2006) are of great interest both for evaluating its thermodynamic characteristics and for optimizing the synthesis of this compound.

Several experimental and theoretical methods were studied to determine the molar heat capacity for CdTe. Experimentally, the molar heat capacities of CdTe were measured by Demidenko (1969) in the 55 K to 300 K-temperature range. Birch (1975) determined the molar heat capacity C_p of CdTe in the temperature range from 1.748 K to 24.82 K. Pashinkin et al. (2002) measured the molar specific heat of ZnTe, CdSe, and CdTe on a DSM-2M calorimeter at 370-640 K (ZnTe) and 500-760 K (CdSe, CdTe). Adachi (2004) determined the heat capacity C_v of CdTe in the 7 K to 700K-temperature range.

On the theoretical side, there have been a few attempts to calculate the molar specific heat. In 2016, Koç and Eser calculated the molar specific heat of CdTe semiconductor by the Debye's model using the the binomial coefficient in the 300-1400K temperature range. Aldeeb (2017) determined the molar specific heat of CdTe semiconductor by the Debye's model depending on Delaney's solution for temperature range of 7-700 K. In addition, Ilchuk et al. (2022) calculated the thermodynamic properties of CdTe crystal, represented in free energy (F), enthalpy (E), entropy (S), specific heat capacity (C_v), and Debye's temperature(θ_D) at 5-1000 K by density function theory (DFT).

This research aims to compare theoretical values calculated by Debye's model with experimental data of molar heat capacity C_v for cadmium telluride (CdTe).

Debye's Model of Heat Capacity C_v

The Debye's model for the temperature dependence of molar heat capacity in a solid at a constant volume (C_v) can be expressed as (Ashcroft and Mermin, 1976; Moustafa, 2007; Allhyani, 2007):

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

Where Z is the number of atoms of one primitive cell, T is the absolute temperature, N_A is the Avogadro number, k_B is the Boltzmann constant, ω_D is the Debye's frequency, ω is the angular frequency, and $\hbar = h/2\pi$, where h is Planck's constant (Ashcroft and Mermin, 1976; Moustafa, 2007; Allhyani, 2007).

The above integral equations are difficult to integrate mathematically, but the mathematician Delaunay succeeded to solve it using a numerical method. Through mathematical substitutions and putting $\omega = P\omega_D$, Delaunay reduced equation (1) to the following form (Delaunay, 1956):

$$d\omega = \omega_D dP \omega = 0 \to \omega_D \quad then \quad P = 0 \to 1$$

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

Where: $k_B\theta_D = \hbar\omega_D, \theta_D$ is the Debye's temperature of solids, and

$R = N_A k_B, R = 8.3144\, J\, mol^{-1} K^{-1}$ is the molar gas constant.

Putting $x = T/\theta_D \& y = C_v/3ZR$, 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 by parts, we get:

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

Since $y = C_v/3ZR$, and for binary compounds (Z = 2), the heat capacity becomes:

$$C_v = 6R\left[\frac{12}{x}\int_0^1 \frac{P^3 dP}{e^{P/x} - 1} - \frac{3}{x}\left(\frac{1}{e^{1/x} - 1}\right)\right], \qquad (5)$$

Results and Discussions

The Debye's model was used to determine the theoretical values of the molar heat capacity (C_v), equation (5) as a function of the absolute temperature (T) and for several Debye's temperatures, 140 K (Strauss, 1977), 144.67 K (Aldeeb, 2017), 160 K (Weber, 2003). MatLab software were used to perform the calculations in the temperature range between 7 and 700 K. Experimental measurements of heat capacity at a constant volume (C_v) for cadmium telluride (CdTe) evaluated by Adachi (2004) are summarized in Table (1).

Table (1): Experomental Data of C_v for CdTe.

T [K]	(Adachi, 2004) $C_{v,exp}\left[\frac{J}{mol.K}\right]$	T[K]	(Adachi, 2004) $C_{v,exp}\left[\frac{J}{mol.K}\right]$
7	0.3859	92	44.3287
14	3.1328	100	45.1256
21	9.5182	142	47.444
28	17.251	180	48.3472
36	24.9688	213	48.7806
43	30.1612	284	49.26
57	37.042	355	49.4847
64	39.292	426	49.6071
71	41.027	497	49.681
78	42.3839	568	49.729
85	43.4615	700	49.783

The following Tables and Figures illustrate the obtained results of the theoretical heat capacities compared with the experiment data (Adachi, 2004).

Table (2) and Figure (1) ($\theta_D = 140$ K), show the convergence between the theoretical results according to Debye's model and the experimental data of heat capacity, especially at medium and high temperatures. At low temperatures there are relative deviations, this is shown by the measure of deviations from the experimental values, which are $5.371\ [\mathrm{J}/(\mathrm{mol}\cdot\mathrm{K})]^2$.

Table (2): Theoretical Values of C_v at $\theta_D = 140$ K Compared with Experimental Data.

T [K]	(Adachi, 2004) $C_{v,exp}\left[\frac{J}{mol.K}\right]$	(Strauss, 1977) $C_{v,theo}\left[\frac{J}{mol.K}\right]$	$C_{v,exp} - C_{v,theo}$	$(C_{v,exp} - C_{v,theo})^2$
7.000	0.38590000	0.48593548809224	0.10003548809224	0.01000709887785
14.000	3.13280000	3.78246964028354	0.64966964028354	0.42207064150615
21.000	9.51820000	10.62551734399865	1.10731734399865	1.22615170032022
28.000	17.25100000	18.39002351454087	1.13902351454087	1.29737456667704

36.000	24.96880000	25.93127801594925	0.96247801594924	0.92636393118559
43.000	30.16120000	30.94993915725716	0.78873915725716	0.62210945819074
57.000	37.04200000	37.56716122341276	0.52516122341276	0.27579431057639
64.000	39.29200000	39.72604252056566	0.43404252056565	0.18839290965899
71.000	41.02700000	41.38989949622212	0.36289949622212	0.13169604435827
78.000	42.38390000	42.69156239452352	0.30766239452353	0.09465614900395
85.000	43.46150000	43.72484446292850	0.26334446292849	0.06935030615510
92.000	44.32870000	44.55640005395518	0.22770005395518	0.05184731457119
100.000	45.12560000	45.32054624088009	0.19494624088009	0.03800403683328
142.000	47.44400000	47.54397189161859	0.09997189161859	0.00999437911380
180.000	48.34720000	48.40991118135677	0.06271118135677	0.00393269226716
213.000	48.78060000	48.82565105041280	0.04505105041280	0.00202959714330
284.000	49.26000000	49.28591011708759	0.02591011708759	0.00067133416749
355.000	49.48470000	49.50104472871708	0.01634472871709	0.00026715015684
426.000	49.60710000	49.61847065132116	0.01137065132116	0.00012929171147
497.000	49.68100000	49.68946717986809	0.00846717986809	0.00007169313492
568.000	49.72900000	49.73562434393320	0.00662434393320	0.00004388193255
700.000	49.78300000	49.78720069403939	0.00420069403939	0.00001764583041
The sum of squares of the deviations = 5.370976133373 $\left(\frac{J}{mol.K}\right)^2$				

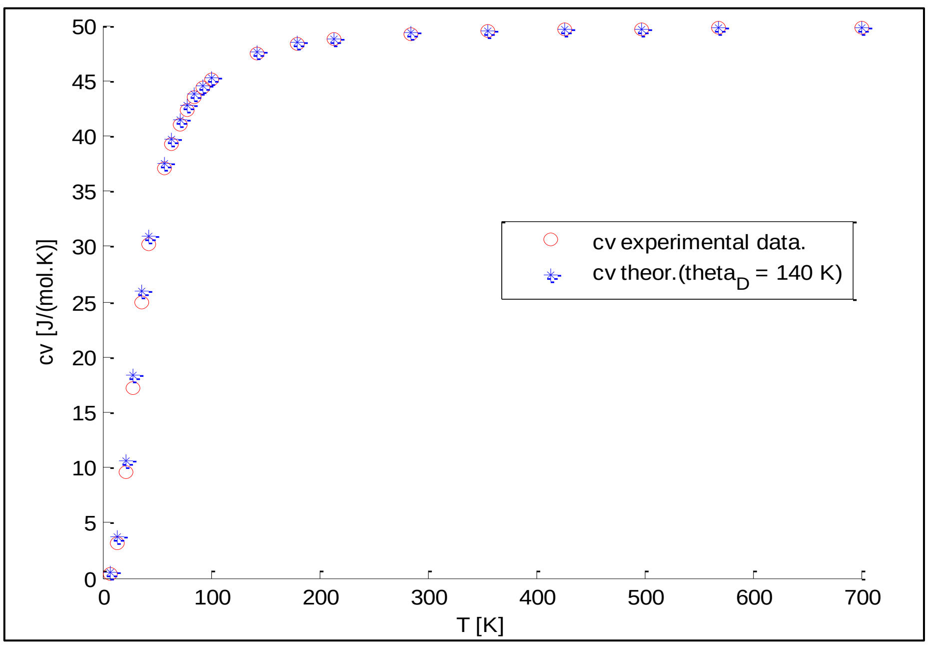

Figure (1): Theoretical Data of C_v Compared with Experimental Measurements.

For the Debye's temperature ($\theta_D = 144.67$ K), Table (3) and Figure (2) show the relationship between the theoretical results according to Debye's model and the experimental data of heat capacity. There is a great convergence between the obtained results. This is shown by the measure of deviations from the experimental values, which are $0.3824\ [\mathrm{J}/(\mathrm{mol}\cdot\mathrm{K})]^2$.

Table (3): Theoretical Values of C_v at $\theta_D = 144.67\ K$ Compared with Experimental Data.

T [K]	(Adachi, 2004) $C_{v,exp}\left[\frac{J}{mol.K}\right]$	(Aldeeb, 2017) $C_{v,theo}\left[\frac{J}{mol.K}\right]$	$C_{v,exp} - C_{v,theo}$	$\left(C_{v,exp} - C_{v,theo}\right)^2$
7.000	0.38590000	0.44038885	0.05448885028306	0.00296903480517
14.000	3.13280000	3.44651840	0.31371840262455	0.09841923614530
21.000	9.51820000	9.87472728	0.35652728306404	0.12711170356903
28.000	17.25100000	17.42449887	0.17349886955431	0.03010185773662
36.000	24.96880000	24.95785757	-0.01094242524124	0.00011973667016
43.000	30.16120000	30.06359167	-0.09760832729383	0.00952738555710
57.000	37.04200000	36.89571493	-0.14628506876266	0.02139932134290
64.000	39.29200000	39.14767364	-0.14432635773312	0.02083009753651
71.000	41.02700000	40.89062604	-0.13637395753514	0.01859785629380
78.000	42.38390000	42.25854251	-0.12535748868843	0.01571449997027
85.000	43.46150000	43.34711126	-0.11438874171846	0.01308478423193
92.000	44.32870000	44.22487105	-0.10382894977776	0.01078045081195
100.000	45.12560000	45.03280874	-0.09279125662209	0.00861021730551
142.000	47.44400000	47.39081938	-0.05318062152738	0.00282817850604
180.000	48.34720000	48.31202310	-0.03517690266919	0.00123741448140
213.000	48.78060000	48.75485654	-0.02574346240576	0.00066272585664
284.000	49.26000000	49.24553260	-0.01446740029665	0.00020930567134
355.000	49.48470000	49.47503635	-0.00966364606908	0.00009338605535
426.000	49.60710000	49.60034597	-0.00675403444889	0.00004561698134
497.000	49.68100000	49.67612293	-0.00487706611130	0.00002378577385
568.000	49.72900000	49.72539364	-0.00360636447302	0.00001300586471
700.000	49.78300000	49.78455431	-0.00254569118013	0.00000648054358
The sum of squares of the deviations = 0.382386081710$\left(\frac{J}{mol.K}\right)^2$				

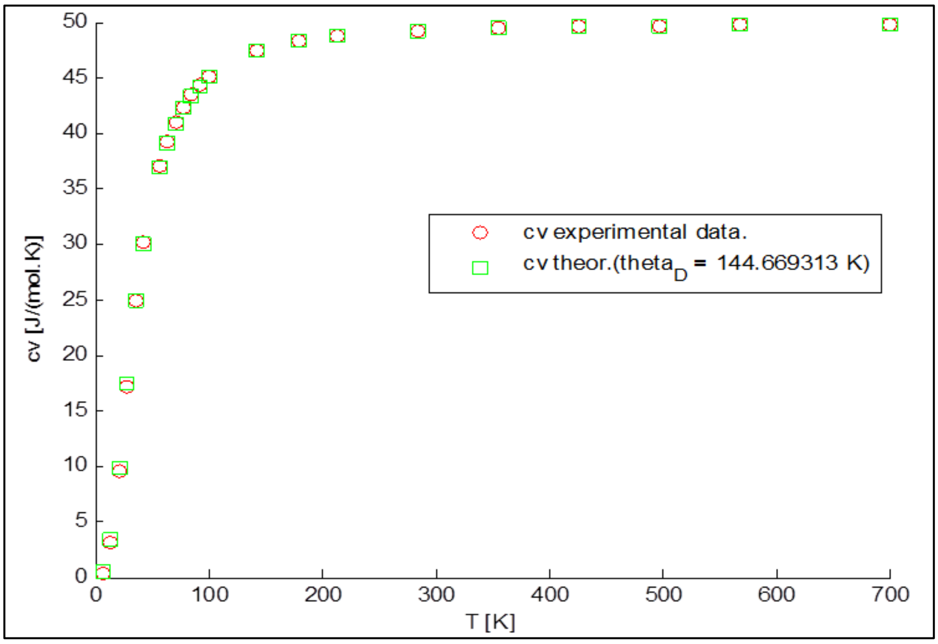

Figure (2): Theoretical Data of C_v Compared with Experimental Measurements.

Table (4) and Figure (3) show the relationship for the Debye's temperature ($\theta_D = 160$ K). There is a large deviation between the theoretical results according to Debye's model and the experimental data of heat capacity, where the value of the deviations to be $49.5827\ [\mathrm{J/(mol \cdot K)}]^2$ .The convergence was only in high-temperatures.

Table (4): Theoretical Values of C_v at $\theta_D = 160\ K$ Compared with Experimental Data.

T [K]	(Adachi, 2004) $C_{v,exp} \left[\frac{J}{mol.K}\right]$	(Waber, 2003) $C_{v,theo} \left[\frac{J}{mol.K}\right]$	$C_{v,exp} - C_{v,theo}$	$(C_{v,exp} - C_{v,theo})^2$
7.000	0.38590000	0.32554342	-0.06035657596861	0.00364291626265
14.000	3.13280000	2.57719315	-0.55560685437555	0.30869897662910
21.000	9.51820000	7.78539282	-1.73280717571837	3.00262070822107
28.000	17.25100000	14.56266512	-2.68833487553684	7.22714440302768
36.000	24.96880000	21.92672655	-3.04207344970084	9.25421087337475
43.000	30.16120000	27.22292258	-2.93827742208857	8.63347420915546
57.000	37.04200000	34.66799398	-2.37400601890000	5.63590457777342
64.000	39.29200000	37.20824159	-2.08375841400805	4.34204912794935
71.000	41.02700000	39.20295407	-1.82404592935897	3.32714355241104
78.000	42.38390000	40.78577466	-1.59812534351687	2.55400461359091
85.000	43.46150000	42.05616033	-1.40533967254004	1.97497959521494
92.000	44.32870000	43.08745830	-1.24124170255609	1.54068096416434
100.000	45.12560000	44.04216623	-1.08343377088440	1.17382873589279
142.000	47.44400000	46.85820362	-0.58579638416577	0.34315740370170

180.000	48.34720000	47.97028245	-0.37691754597098	0.14206683646079
213.000	48.78060000	48.50724734	-0.27335266321316	0.07472167848573
284.000	49.26000000	49.10402186	-0.15597813588226	0.02432917887330
355.000	49.48470000	49.38379866	-0.10090133865444	0.01018108014226
426.000	49.60710000	49.53673152	-0.07036847821029	0.00495172272563
497.000	49.68100000	49.62927238	-0.05172761778272	0.00267574644148
568.000	49.72900000	49.68946718	-0.03953282018430	0.00156284387172
700.000	49.78300000	49.75675814	-0.02624185756298	0.00068863508836
The sum of squares of the deviations = 49.58271838 $\left(\frac{J}{mol.K}\right)^2$				

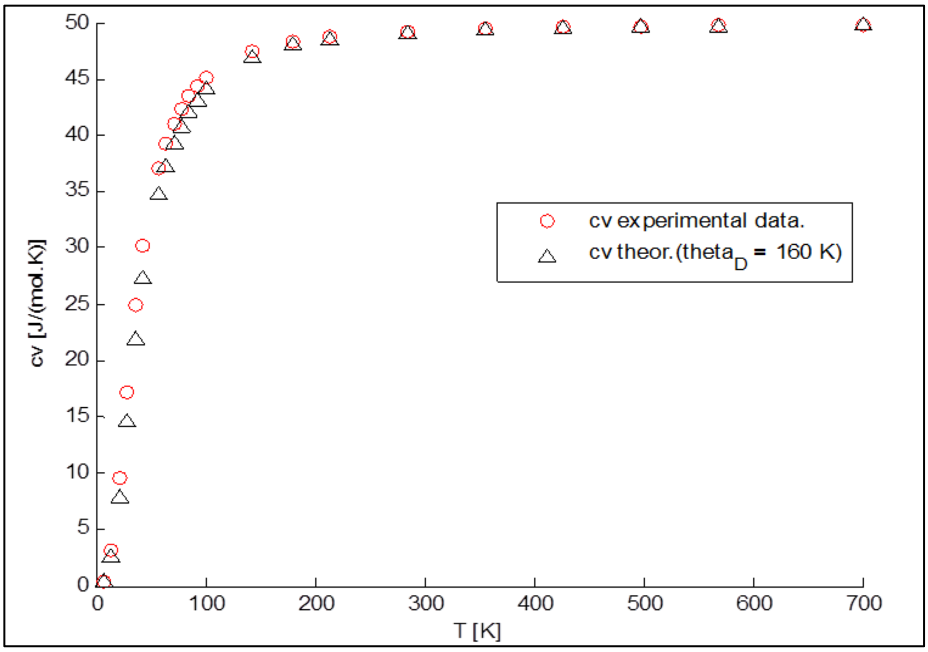

Figure (3): Theoretical Data of C_v Compared with Experimental Measurements.

Figure (4) plots the curves showing comparison experimental data (Adachi, 2004) with calculated theoretical values using the three Debye's temperatures. They are greatly compatible with the experimental data in medium and high temperatures, but there is a deviation between the calculated values and the experimental data at low temperatures. At the Debye's temperature of 144.67 K was the best fit between the experimental and the theoretical results almost in the entire temperatures range. This is evident from the values of the sum of squared deviations where it was $0.3824\ [J/(mol \cdot K)]^2$.

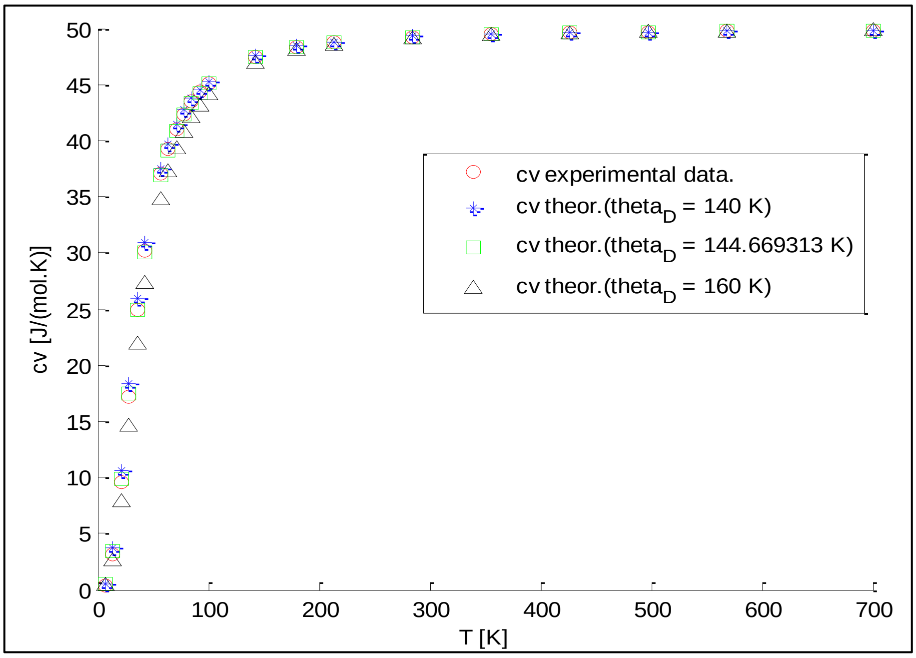

Figure (4): Comparison Experimental Data with Calculated Theoretical Values using the Three Debye's Temperatures.

Conclusion

In this research work, the Debye's model was used to calculate the molar heat capacity C_v for the purpose of estimating a Debye's temperature for CdTe. The experimental measurements were compared with the calculated values using several Debye temperatures $(\theta_D = 140\ \mathrm{K}, 144.67\ \mathrm{K} \& \ 160\ \mathrm{K})$ to determine the best congruence between the calculated and the experimental results of heat capacity C_v. The results have shown that the calculated theoretical values of C_v greatly compatible with the experimental data in medium and high temperatures. The best fit between the experimental and the theoretical results was almost in the entire temperature range for the Debye's temperature of 144.67K. This is evident from the values of the sum of squared deviations where it was $0.3824\ [\mathrm{J}/(\mathrm{mol} \cdot \mathrm{K})]^2$.

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