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Thermodynamic Properties of Tantalum

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Abstract The thermodynamic properties of tantalum have been evaluated to 5800 K. Selected values include an enthalpy of sublimation of 781 ± 4 kJ/mol at 298.15 K and a boiling point at one atmosphere pressure of 5762 K.

Keywords gas · liquid · solid · tantalum · thermodynamic properties

1 Introduction

This work is one of a series on thermodynamic properties of the elements to appear in the *Journal of Phase Equilibria and Diffusion*. The articles are expected to be especially useful to those who are updating databases for phase diagram prediction and for other applications as well. Each article in the series will list the properties of a single element, including Ag,[1] Au,[2] Be,[3] Cr, Cu,[4] Hf,[5] Mo, Nb,[6] Ta, Ti, V[7] and W. References are given for papers which have already been published. In addition, the series will also include two summary papers on selected values of the melting points and boiling points of the elements and on the enthalpies and entropies of fusion and transition of the elements.

Inaba[8] determined the superconducting temperature to be 4.4924 K whilst a melting point of 3293 ± 15 K is selected as an average after correction to ITS-90 of values of 3292 K determined by Rudy and Harmon[9] and 3294 K determined by Pemsler[10] where in both cases the material used was of particularly high purity. Although Hiernaut et al.[11] initially also reported a similar value at 3290 K, later Hiernaut et al.[12] reported a much lower value of 3260 K without giving any explanation as to the large difference. Cezairliyan et al.[13] also selected a much lower melting point of 3258 K as a correction to ITS-90 of the value obtained by Malter and Langmuir[14] although no impurity content was given so that this value must be treated with caution. Rudy and Harmon[9] noted that increasing the total impurity content to only 500 ppm (0.05%) resulted in a 30 K drop in the melting point.

Wherever possible values have been corrected to the currently accepted atomic weight of 180.94788 ± 0.00002[15] and to the ITS-90 temperature scale using correction factors of Douglas,[16] Rusby,[17] Rusby et al.[18] and Weir and Goldberg.[19]

Based on dilatometric measurements, Bollinger et al.[20] suggested that all of the three Group 5 elements, vanadium, niobium and tantalum, underwent a martensitic structural distortion just below room temperature leading to a structure which was not cubic. However, although given in smooth form, neither specific heat measurements of Sterrett and Wallace[21] (12-543 K) or electrical resistivity measurements by Williams et al.[22] (80-400 K) showed any evidence of an anomaly that would be involved in a phase transition whilst lattice parameter measurements of Smirnov and Finkel'[23] established that the structure remained body-centered cubic down to at least 127 K. Therefore, until the observations of Bollinger et al.[20] are independently verified and the supposed new low temperature structure is fully characterized then it is assumed that the body-centered cubic structure exists over the whole temperature range (Fig. 1, 2, and 3).

Previous comprehensive reviews on tantalum were by Hultgren et al.,[24] Gurvich et al.[25] and JANAF (Chase[26]) and on the solid only by Maglić[27] and Bodryakov.[28]

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Fig. 1 Low temperature specific heat of solid tantalum, taken from Table 15

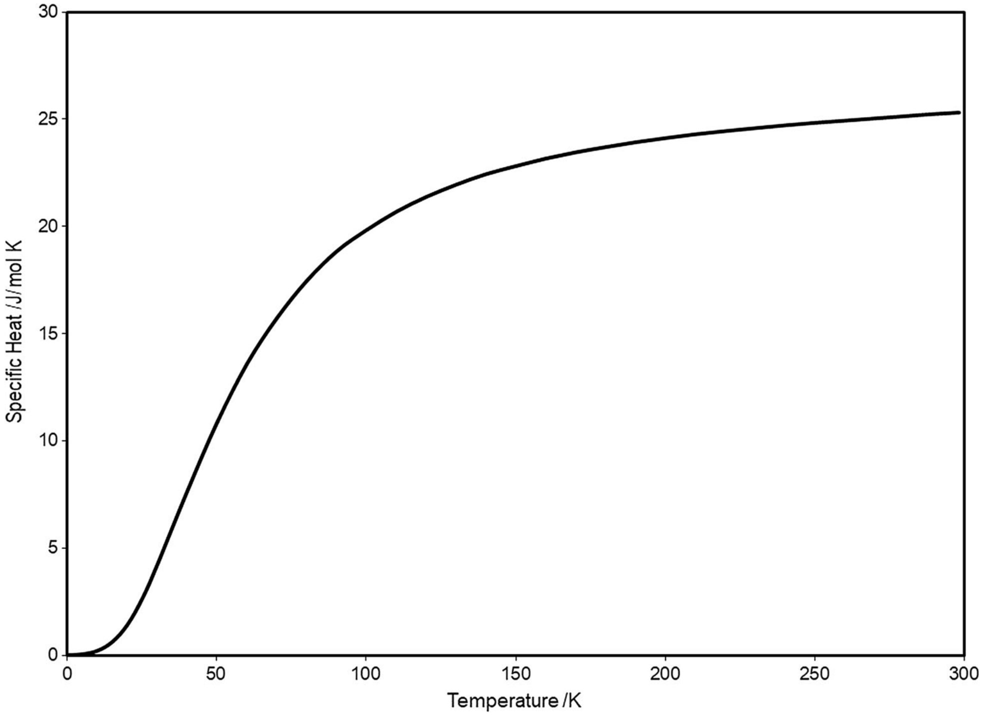
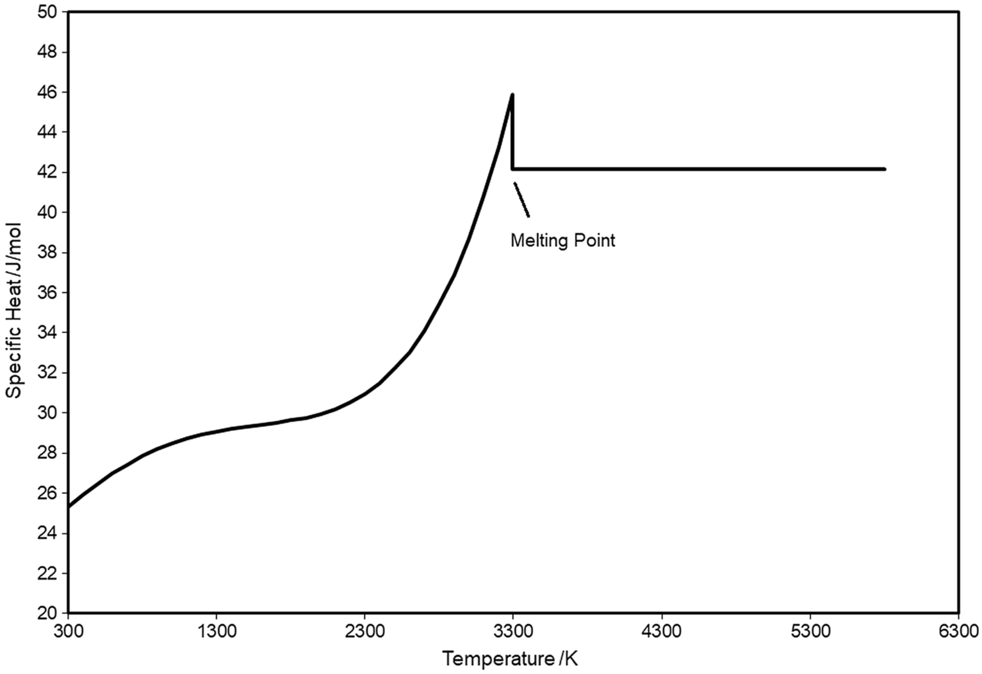


Fig. 2 Specific heat of tantalum for 300 < T < 5800 K, taken from Table 16



2 Solid Phase

2.1 Superconducting State—Range 0 to 4.4924 K

In the description of the electronic structure of transition metals in terms of two electronic conduction bands then if the scattering of electrons between the two bands is sufficiently weak then both may separately undergo a transition into the superconducting state with the resultant formation of two energy gaps which is a Type II superconductor. Anomalies which appeared to confirm this behaviour in niobium were summarised by Sellers et al.[29] whilst, in particular, a similar anomaly was observed for tantalum in the specific heat measurements of Satoh et al.[30] However, based on specific heat measurements the anomalies for niobium were shown by Sellers et al.[31] to be due to

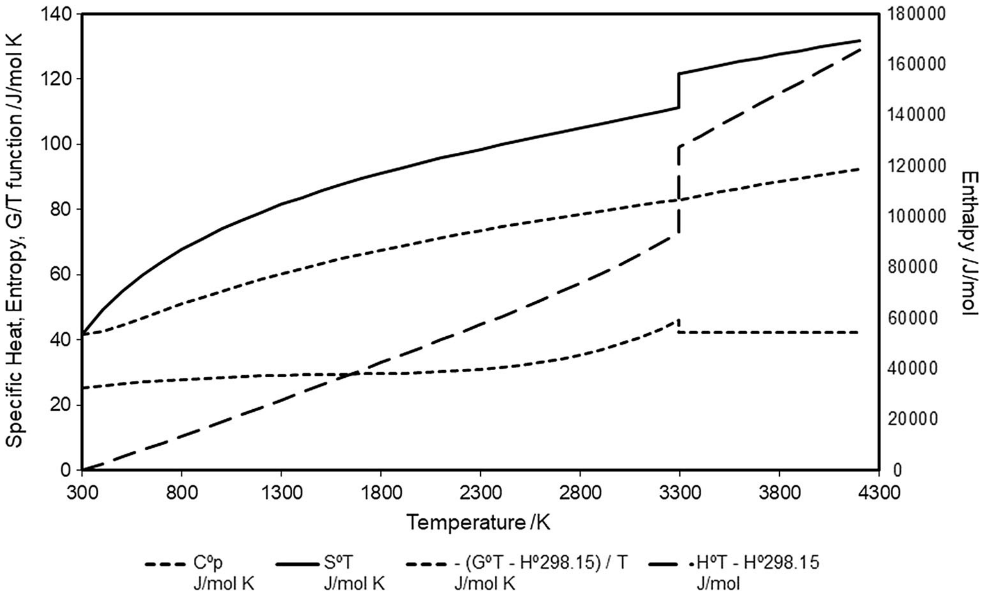


Fig. 3 High temperature thermodynamic properties of tantalum for $300 < T < 4300$ K, taken from Table 16

Table 1 Low temperature normal state specific heat constants

Authors	Ref	γ_1 mJ/(mol K^2)	θ_{D1} K	γ_2 mJ/(mol K^2)	θ_{D2} K
Keesom and Desirant	69	5.90	246.5	...	...
Worley et al.	70	5.44	231	...	...
Wolcott	71	5.87	245	...	...
Chou et al.	72	5.69*	248 *	3.35 *	222 *
White et al.	73	5.69	255	...	...
Morin and Maita	74	6.28	240	...	...
Corzan and Cook	75	5.85	251	...	...
Sellars et al.	29	6.2	...	...	...
Shen	32	6.02	258.2	...	...
Leupold et al.	33	5.42	238.0	4.36	227.6

*As recalculated by Leupold et al.[33]

hydrogen contamination and that with hydrogen removed niobium behaved as a typical Type I superconductor. On this basis, specific heat measurements of Shen[32] were selected for tantalum and below $T_c/2$, were T_c is the superconducting transition temperature, the electronic contribution to the specific heat (C_{es}) was fitted to the Bardeen–Cooper–Schrieffer equation which together with two lattice contributions leads to the full representation of the specific heat ($C_{s)}$ below 2.25 K ($T_c/2$) as given by Eq 1:

$$\begin{aligned} C_s(\mathrm{mJ/mol\ K}) = C_{es} + AT^3 + BT^5 = 8.13\gamma T_c \\ \times \exp(-1.45\ T_c/T) + 0.113\ T^3 \\ + 1.9 \times 10^{-4} T^5 \end{aligned} \qquad \text{(Eq 1)}$$

where γ is the normal state electronic coefficient (6.02 mJ/mol K^2) and T_c is selected as a working value of 4.467 K as an average of values determined for the samples I and II of Shen.

At T_c, a value of $(C_s - C_{n)}/\gamma\ T_c = 1.53$ is selected as an average of samples I and II of Shen[32] and when extrapolated to the selected value of T_c leads to $C_s - C_n = 41.37$ mJ/(mol K) so that $C_s = 79.01$ mJ/(mol K) at T_c. This value was combined with the value of 13.59 mJ/(mol K) at 2.25 K obtained from Eq 1 and an intermediate value of at the mid-point temperature of 3.369 K obtained by iteration so that at T_c the entropy values of the

Table 2 Selected values for the solid at 298.15 K

Authors	Ref	$C°_p$ J/(mol K)	$H°_{298.15\ K} - H°_0$ J/mol	$S°_{298.15\ K}$ J/(mol K)
Hultgren et al.	24	25.35	5636	41.51
Gurvich et al.	25	25.29	5680	41.52
JANAF (Chase)	26	25.30	5681	41.47
This work	...	25.30	5684	41.56

superconducting state and the normal state are equal. This gives Eq 2 for C_s over the range 2.25 K to T_c:

$$C_s(\mathrm{mJ/mol\ K}) = C_{es} + AT^3 + BT^5 = -9.1652 + 1.7747T + 3.4493T^2 + 0.113T^3 + 1.9 \times 10^{-4}T^5 \quad \text{(Eq 2)}$$

Derived thermodynamic values based on Eq 1 and 2 are given in Table 14 whilst differences from the selected values of other measurements in this region are given in Table 10.

2.2 Normal State—0 to 298.15 K

Low temperature specific heat for tantalum in the normal state is given in terms of the modified Debye equation: $C_p = D/T^2 + \gamma\ T + A\ T^3 + B\ T^5$ where D is the nuclear quadrupole coefficient, γ is the electronic coefficient and A and B are lattice contributions where A is usually represented in terms of a limited Debye temperature, θ_D, where $\theta_D^3 = (12/5)\ \pi^4\ R/A = 1943.770/A$ where R is the Gas Constant and A is given in units of J/(mol K^4). For D, Shen[32] gives the value 0.01 mJ K/mol which is accepted. Leupold et al.[33] showed that discrepancies in the reported values of γ and θ_D could be explained by an abrupt change in the slope of the specific heat values at 7.19 K which was due to the presence of a Kohn anomaly in the phonon density of states. Experimental values of γ and θ_D are summarised in Table 1 where the subscripts γ_1 and θ_{D1} and γ_2 and θ_{D2} distinguish values above and below the transition temperature.

The measurements of Shen[32] and Leupold et al.[33] initially appear to disagree significantly with a preference being given to the values of Shen since the value of θ_D at 258 K gave the best agreement with a value of 263.8 K selected by Alers[34] from elastic constant measurements. However, the two sets of measurements were reconciled as follows to cover the range up to 12.5 K:

$$\text{Shen}: 0\ \text{to}\ 4.718\,K: C_n(\mathrm{mJ/mol\ \ K}) = 0.01/T^2 + 6.02\,T + 0.113\,T^3 + 1.9\,x\,10^{-4}T^5 \quad \text{(Eq 3)}$$

Table 3 Enthalpy of fusion and liquid specific heat values for tantalum

Authors	Ref	ΔH s, l kJ/mol	$C°_p$ J/(mol K)
Lebedev et al.	76	36.6	...
Martynyuk and Tsapkov	77	33.0	...
Lebedev and Mozharov	78	37.5	...
Shaner et al.	79	36.2	68.8
Gathers	80	36.2	59.2
Gallob et al.	81	41.6	44.3
Berthault et al.	82	32.0	38.0
Jäger et al.	83	27.1	45.2
Hixson	84	25.3	46.3
McClure and Cezailiyan	52	34.8 ± 2.1	...
Pottlacher and Seifter	53, 54	31.5	40.9
Arpaci and Frohberg	37	33.89 ± 0.22	42.18 ± 4.62
Selected		33.883 ± 0.402	42.18 ± 4.62

$$\text{Leupold et al.}: 4.718\ \text{to}\ 7.19\,\text{K}: C_n(\mathrm{mJ/mol\ \ K}) = 5.42\,T + 0.144183\,T^3 \quad \text{(Eq 4)}$$

$$\text{Leupold et al.}: 7.19\ \text{to}\ 12.5\,\text{K}: C_n(\mathrm{mJ/mol\ \ K}) = 4.36\,T + 0.164865\,T^3 \quad \text{(Eq 5)}$$

Above 12.5 K and up to 298.15 K the specific heat measurements of Sterrett and Wallace[21] (12 to 543 K) were selected since these give the best agreement with the high temperature selected values. Because the values were given in a smooth form they were not separately fitted to analytical equations. Laser flash calorimetric measurements of Takahashi and Nakamura[35] (82 to 997 K) agree satisfactorily in the low temperature region trending from 0.1% high at 90 K to 0.7% high at 150 K to 0.4% low at 298.15 K. Thermodynamic values in the normal state below the superconductive transition temperature are also included in Table 14 whilst thermodynamic values at 5 K and above are given in Table 15. The deviations of other specific heat values in the low temperature region are given in Table 11.

2.3 Solid State: 298.15 to 3293 K

The requirement is not only to have a continuity with the low temperature specific heat curve but also to agree with the closely agreeing high temperature enthalpy values of Berezin and Chekhovskoi[36] (2440 to 3238 K) and Arpaci and Frohberg[37] (2653 to 3276 K in the solid range) in order to derive an enthalpy of fusion value which agrees with experimental values.

Table 4 Energy levels of tantalum gas phase

Authors	Ref	Number of Levels
Moore	59	260
Guthöhrlein et al.	85	1
Messnarz and Guthöhrlein	86	35
Arcimowicz et al.	87	10
Jaritz et al.	88	14
Messnarz et al.	89	44
Jaritz et al.	90	18
Jaritz et al.	61	28
Jaritz et al.	91	43
Glowacki et al.	92	35

Specific heat measurements of Rasor and McClelland[38,39] (700 to 3200 K), Lehman[40] (100 to 3200 K) and Taylor and Finch[41,42] (100 to 3195 K) all showed a sudden sharp increase above 2800 K which was not shown in the measurements of Cezairliyan et al.[43] (1900 to 3200 K). Whilst attempts have been made to explain the sharp increase as real behaviour it is considered that it is simply due to sample oxidation or experimental error as evidenced by a similar sudden sharp increase observed in the complimentary thermal expansion measurements of Rasor and McClelland[38,39] when compared to the quality measurements of Petukov et al.[44] (1244 to 2267 K) and Miiller and Cezairliyan[45,46] (1500 to 3200 K). On these grounds, and because the first three sets of measurements differ significantly from those of Cezairliyan et al.,[43] then it is considered that the measured high temperature specific heat values of tantalum are unsatisfactory for representing the thermodynamic properties and therefore instead values are obtained by combining the agreeing drop calorimetry enthalpy measurements of Oetting and Navratil[47] (534 to 1383 K) (8 data points), Berezin and Chekhovskoi[36] (2440 to 3238 K) (11 data points out of 18) and Arpaci and Frohberg[37] (2653 to 3276 K in the solid state) (15 data points out of 19). The first two sets of measurements were

Table 5 Enthalpies of sublimation at 298.15 K

Authors	Ref	Method	Temperature range K	$\Delta H^\circ_{298.15}$ (II) kJ/mol	$\Delta H^\circ_{298.15}$ (III) kJ/mol	Notes
Babeliowsky	93	MS	2578-3042	705 ± 17	...	(a)
Zandberg et al.	94	MS	2400-2660	689 ± 29	...	
Sasaki et al.	95	MS	2550-2770	774 ± 12	...	
Langmuir and Malter	62	L	1997-3253	765 ± 1	777.7 ± 0.8	
Fiske	63	L	2630-2846	630 ± 43	761.3 ± 1.8	
Edwards et al.	65	L	2624-2948	792 ± 12	780.3 ± 0.5	
Gebhardt et al.	96	Evap	2900-3150	972 ± 16	794.7 ± 1.8	
Gorbatyi and Shuppe	66	Evap RT	2070-2400	768 ± 6	779.9 ± 0.3	
Golubtsov and Nesmeyanov	67	Evap RT	1875-2970	789	788.4 ± 0.1	(b)
		Eff RT	2389-2725	784	783.8 ± 0.1	(b)
Selected					781 ± 4	

$\Delta H^\circ_{298.15}$ (II) and $\Delta H^\circ_{298.15}$ (III) ar the Second Law and Third Law enthalpies of sublimation at 298.15 K

(a)Value given only at 298.15 K

(b)Values given only in the form of the Clausius–Clapeyron equation

Methods for Table 5

Eff RT effusion with radioactive tracer, *Evap RT* evaporation with radioactive tracer

Evap evaporation, *L* Langmuir method, *MS* mass spectrometry

Table 6 Vapour Pressure Equations

Phase	Range K	A	B	C	D	E
Solid	1800-3293	48.83554	− 4.53932	− 96587.05	2.31330×10^{-3}	-1.80086×10^{-7}
Liquid	3293-5800	27.48068	− 1.34980	− 93376.75	5.08569×10^{-5}	4.01934×10^{-9}

Table 7 High temperature representative equations

Solid: 298.15 to 3293 K

$C^\circ_p(\text{J/mol K}) = 23.2424 + 7.35950 \times 10^{-3}T - 1.086909 \times 10^{-6}T^2 - 1.605216 \times 10^{-9}T^3 + 5.74080 \times 10^{-13}T^4$

$H^\circ_T - H^\circ_{298.15K}(\text{J/mol}) = 23.2424T + 3.67975 \times 10^{-3}T^2 - 3.62303 \times 10^{-7}T^3 - 4.01304 \times 10^{-10}T^4 + 1.14816 \times 10^{-13}T^5 - 7244.32$

$S^\circ_T(\text{J/mol K}) = 23.2424\ln(T) + 7.35950 \times 10^{-3}T - 5.434545 \times 10^{-7}T^2 - 5.35072 \times 10^{-10}T^3 + 1.43520 \times 10^{-13}T^4 - 93.0027$

Liquid: 3293 to 5800 K

$C^\circ_p\ (\text{J/mol K}) = 42.1820$

$H^\circ_T - H^\circ_{298.15\ K}\ (\text{J/mol}) = 42.1820\ T - 11493.9$

$S^\circ_T\ (\text{J/mol K}) = 42.1820\ \ln(T) - 220.0043$

Table 8 Free energy equations above 298.15 K

Solid: 298.15 to 3293 K

$G^\circ_T - H^\circ_{298.15\ K}(\text{J/mol}) = 116.2451\ T - 3.67975 \times 10^{-3}T^2 + 1.811515 \times 10^{-7}T^3 + 1.33768 \times 10^{-10}T^4 - 2.87040 \times 10^{-14}T^5 - 23.2424\ T\ln(T) - 7244.32$

Liquid: 3293 to 5800 K

$G^\circ_T - H^\circ_{298.15\ K}\ (\text{J/mol}) = 262.1863\ T - 42.1820\ T\ \ln(T) - 11493.9$

Table 9 Transition values involved with the free energy equations

Transition	Temperature T K	ΔH° s, l J/mol	ΔS° s, l J/(mol K)
Fusion	3293	33883.093	10.2894

directly corrected to ITS-90 whilst the measurements of Arpaci and Frohberg[37] were corrected to the selected melting point. Enthalpy measurements of Conway and Hein[48,49] (1315 to 2622 K) (12 data points out of 19) were also considered but were then rejected since they showed an average bias of 0.5% low compared to the selected equation. The selected values were fitted to Eq 6 which has an overall accuracy as a standard deviation of ± 310 J/mol (0.40%). The selection of this equation to cover the range from 298.15 K to the melting point gave the best continuity with the selected low temperature specific heat values:

$$H^\circ_T - H^\circ_{298.15\,K}(\text{J/mol}) = 23.2424\,T + 3.67975\,x\,10^{-3}T^2 - 3.62303\,x\,10^{-7}T^3 - 4.01304\,x\,10^{-10}T^4 + 1.14816\,x\,10^{-13}T^5 - 7244.32 \quad \text{(Eq 6)}$$

Derived specific heat and entropy equations are given in Table 7. Thermodynamic properties derived from Eq 6 are given in Table 16 whilst deviations for specific heat and other enthalpy values are given in Tables 12 and 13 respectively.

2.4 A Comparison of Selected Values at 298.15 K

The comparison is given in Table 2. In the low temperature region above the superconducting temperature, Hultgren et al.[24] mainly combined the specific heat values of Sterrett and Wallace[21] and Kelley[50] whilst JANAF

Table 10 Deviations of superconducting specific heat values

Authors	Ref	Temperature range K	% deviations from the selected values
Keesom and Desirant	69	1.4-4.2	Sample 1: Scatters 7.7 high to 35.0 high over range 1.4-3.7 K; Sample 2: Trends from initially 6.6 high to 2.8 high at 3.8 K; Sample 3: Scatters 1.7 high to 8.0 high over range 3.2-3.9 K
Worley et al.	70	1.7-4.4	Trends from initially 6.4 high to 7.0 high at 2.5 K to 5.0 high at 4.4 K
White et al.	73	1.3-4.3	Marked scatter 3.2 high to 60.1 high below 1.5 K. Above 1.5 K averages 3.6 high to 2.1 K then trends to 11.7 high at 2.4 K and falls to 2.4 high at 4.3 K

Table 11 Deviations of low temperature normal state specific heat values

Authors	Ref	Temperature Range K	% deviations from the selected values
Keesom and Desirant	69	1.3-3.4	Scatters 12.7 low to 8.3 high
Worley et al.	70	3.3-5.0	Trends from initially 7.2 low to 3.9 high
White et al.	73	1.4-24.3	In zero magnetic field above 4.5 K trends from initially 0.3 high to 4.5 low at 6.7 K to an average of 7.7 high above 23 K. In magnetic field (1.4 to 5.4 K) trends from initially 3.9 low to 7.7 low at 2.0 K to 0.2 low at 4.3 K to 4.0 low at 5.4 K
Simon and Ruhemann	97	71.1-78.0	Averages 6.4 low
Kelley	50	53-295	Trends from initially 0.3 high to 1.5 high at 75 K falling to 0.1 high at 165 K and then averaging 0.2 high above this temperature
Clusius and Losa	51	10-274	Marked scatter 8.6 low to 0.8 low below 15 K. Above this temperature averages 3.6 low below 25 K then trends to 0.3 high at 51 K to 2.1 low at 88 K to 0.6 high at 274 K.
Lehman	40	100-3200	Scatters 4.0 low to 2.7 low below 300 K
Taylor and Finch	41, 42	100-3195	Trends from initially 4.6 high to 2.7 low below 300 K
Takahashi and Nakamura	35	82-997	In the low temperature region trends from 0.1 high at 90 K to 0.7 high at 150 K to 0.4 low at 298.15 K

(Chase[26]) combined these two sets of measurements with those of Clusius and Losa.[51] Both Gurvich et al.[25] and in the present review preference is given to the measurements of Sterrett and Wallace.[21]

3 Liquid Phase

Enthalpy measurements of Arpaci and Frohberg[37] (3292 to 3358 K in the liquid state) were corrected to the selected melting point and fitted to Eq 7 with an overall accuracy as a standard deviation of ± 256 J/mol (0.18%):

$$H^\circ_T - H^\circ_{298.15\,K}(\text{J/mol}) = 42.1820\,T - 11493.9 \qquad (\text{Eq } 7)$$

The derived enthalpy of fusion at 33.883 ± 0.402 kJ/mol can be compared with the wide range of reported values given in Table 3, all of which, apart from the measurement of Arpaci and Frohberg,[37] were determined by rapid pulse heating. However, it is noted that the directly determined value of McClure and Cezairliyan[52] and the derived value of Pottlacher and Seifter[53,54] satisfactorily agree with the selected value.

Because of the very narrow 65 K range over which the drop calorimetry measurements of Arpaci and Frohberg were measured then the derived liquid specific heat is relatively poorly determined as 42.18 ± 4.62 J/(mol K). Other determinations of liquid specific heat used rapid pulse heating and are also given in Table 3 except for the measurements of Lebedev and Mozharov[55] which trend from 69.6 J/(mol K) at 3300 K to 27.7 J/(mol K) at 3900 K. It is considered that the measurements of Pottlacher and Seifter[53,54] at 40.9 J/(mol K) represents an

Table 12 Deviations of enthalpy values in the solid range

Authors	Ref	Temperature range K	% deviations from the selected values
Magnus and Holzmann	98	574-1173	Initially 1.2 high trending to an average of 0.9 high above 900 K
Jaeger and Veenstra	99	572-1828	Initially 1.5 low trending to 2.7 low at 1074 K and to 0.8 low at 1828 K
Fieldhouse et al.	100	841-1910	Initially scatter from 0.7 high to 14.6 high below 1700 K but above 1700 K scatters 1.4 high to 20.7 high
Hoch and Johnston	101	1362-2847	Initially 2.4 low trending to 1.1 low at 2508 K to 2.6 low at 2847 K
Conway and Hein	48, 49	1315-2622	Scatters 2.2 low to 0.2 high
Shaner et al.	79	2200-3270	Trends from initially 0.3 high to 1.7 high
Pottlacher and Siefter	53	2500-3270	Trends 0.2 low to 4.8 high at 2900 K and an average of 5.5 high at 3000 K and above.

Table 13 Deviations of high temperature solid specific heat values

Authors	Ref	Temperature range K	% deviations from the selected values
Pirani	102	380-1400	Trends from initially 2.7 low to 9.4 low at 700 K to 11.7 high above 1400 K
Langmuir and Malter	103	1656-2800	Trends from initially 4.0 high to 12.5 high at 2400 K to 7.4 high at 2800 K
Sterrett and Wallace	21	12-543	Above 298.15 K trends to 0.3 high at 370 Kto 0.1 high at 420 K to 0.4 high at 543 K
Boggs et al.	104	1089, 1589	5.0 low and 8.1 high
Boggs and Weibelt	105	1089-1700	Sample 1: Trends from initially 10.3 high to 1.9 low at 1255 K to 26.4 high at 1700 K; Sample 2: Trends from initially 4.5 high to 23.2 high at 1422 K to 17.2 high at 1700 K; Sample 3: Trends from initially 3.5 low to 14.7 high at 1422 K to 5.7 high at 1700 K
Lehman	40	100-3200	Above 300 K trends from initially 1.3 low to 2.2 low at 500 K to 16.7 high at 3200 K
Rasor and McClelland	38, 39	700-3200	Trends from initially 3.5 low to 11.3 high at 2800 K to 6.8 high at 3200 K
Taylor and Finch	41, 42	100-3200	Above 300 K trends from initially 1.9 low to 3.5 low at 800 K to 19.7 high at 3195 K
Hildenbrand et al.	106	1200-1700	Trends from initially 16.2 low to 6.6 low at 1600 K to 1.0 high at 1700 K
Lowenthal	107	1200-2400	Trends from initially 3.2 low to 4.8 high at 2400 K
Kraftmakher	108, 109	1200-2900	Trends from initially 4.4 low to 0.7 high at 2200 K to 5.3 low at 2900 K
Affortit	110	600-3100	Trends from initially 16.3 low to 18.4 low at 900 k to 6.3 high at 2400 K to 0.5 low at 3100 K to 2.5 high at 3200 K
Cezairliyan et al.	43	1900-3200	Sample 1 and also samples 3 and 4 above 3000 K: Trends from initially 3.0 high to 6.2 high at 2400 K to 1.4 high at 3100 K and 2.8 high at 3200 K
Filippov and Yurchak	111, 112	1100-2900	Sample 1: Trends from initially 1.7 low to 2.1 high at 2100 K to 1.4 high at 2300 K; Sample 2: Trends from initially 2.9 low to 3.2 high at 2100 K to 2.0 high at 2400 K; Sample 3: Trends from initially 0.6 high to 5.0 high at 2200 K to 6.5 low at 2900 K
Lebedev and Mozharov	55	2300-3200	Trends from initially 3.4 high to 4.1 high at 2600 K then to 1.1 high at 2900 K and 59.5 high at 3200 K
Kulish and Filippov	113	300-1300	Trends from initially 1.6 low to 3.3 low at 500 K to 1.7 low at 1300 K
Gilchrist and Preston	114	323-973	Trends from initially 1.3 high to 3.5 low at 673 K to 2.6 low at 973 K
Milošević et al.	115	300-2300	Trends from initially 1.4 high to 3.1 low at 1100 K to 8.4 high at 2300 K
Takahashi and Nakamura	35	82-997	In the high temperature region trends from 0.4 low at 298.15 K to 1.1 low at 700 K to 0.4 high at 997 K

Table 14 A comparison of thermodynamic properties below the superconducting temperature

T K	Superconducting state			Normal state		
	C°_s mJ/(mol K)	$H^\circ_T - H^\circ_0$ mJ/mol	S°_T mJ/(mol K)	C°_n mJ/(mol K)	$H^\circ_T - H^\circ_0$ mJ/mol	S°_T mJ/(mol K)
0.5	0.0146	0.0017	0.0042	3.064	0.754	3.015
1.0	0.450	0.0658	0.0788	6.143	3.038	6.058
1.5	3.297	0.861	0.686	9.417	6.916	9.157
2.0	9.487	3.931	2.409	12.95	12.49	22.34
2.5	18.61	10.82	5.445	16.84	19.92	15.64
3.0	30.30	22.95	9.838	21.16	29.40	19.09
3.5	44.24	41.49	15.53	26.02	41.17	22.70
4.0	60.55	67.59	22.47	31.51	55.52	26.53
4.4924	79.01	101.85	30.53	37.64	72.51	30.53

Table 15 Low temperature thermodynamic properties

Temperature K	C°_p J/(mol K)	$H^\circ_T - H^\circ_0$ J/mol	S°_T J/(mol K)	$-(G^\circ_T - H^\circ_0)$ J/mol	$-(G^\circ_T - H^\circ_0)/T$ J/(mol K)
5	0.0451	0.123	0.0349	0.0519	0.0104
10	0.208	0.676	0.105	0.378	0.0378
15	0.623	2.624	0.258	1.243	0.0829
20	1.415	7.540	0.535	3.164	0.158
25	2.641	17.53	0.976	6.870	0.274
30	4.211	34.61	1.595	13.22	0.441
40	7.634	93.76	3.272	37.14	0.928
50	10.849	186.7	5.333	79.95	1.599
60	13.582	309.3	7.561	144.3	2.406
70	15.717	456.4	9.823	231.3	3.304
80	17.454	622.5	12.039	340.7	4.258
90	18.831	804.3	14.179	471.8	5.242
100	19.827	997.7	16.216	623.9	6.239
110	20.681	1200	18.146	795.8	7.234
120	21.372	1410	19.976	986.5	8.221
130	21.941	1627	21.710	1195	9.192
140	22.435	1849	23.354	1420	10.146
150	22.807	2075	24.915	1662	11.079
160	23.159	2305	26.398	1918	11.990
170	23.452	2538	27.811	2190	12.880
180	23.695	2774	29.159	2474	13.747
190	23.921	3012	30.446	2772	14.592
200	24.113	3252	31.678	3083	15.416
210	24.293	3494	32.859	3406	16.218
220	24.440	3738	33.992	3740	17.001
230	24.578	3983	35.082	4086	17.763
240	24.707	4230	36.138	4442	18.507
250	24.825	4477	37.142	4808	19.232
260	24.925	4726	38.117	5184	19.940
270	25.030	4976	39.060	5570	20.631
280	25.134	5227	39.972	5965	21.305
290	25.235	5478	40.856	6370	21.964
298.15	25.302	5684	41.556	6705	22.490

Table 16 High temperature thermodynamic properties

Temperature K	C°_p J/(mol K)	$H^\circ_T - H^\circ_{298.15}$ J/mol	S°_T J/(mol K)	$-(G^\circ_T - H^\circ_{298.15})/T$ J/(mol K)
298.15	25.302	0	41.556	41.556
300	25.314	46.8	41.713	41.556
400	25.924	2609	49.080	42.557
500	26.486	5230	54.926	44.466
600	26.994	7904	59.800	46.626
700	27.449	10,627	63.996	48.815
800	27.848	13,392	67.688	50.948
900	28.192	16,195	70.989	52.995
1000	28.484	19,029	73.975	54.946
1100	28.727	21,890	76.701	56.801
1200	28.925	24,773	79.201	58.565
1300	29.086	27,674	81.531	60.244
1400	29.216	30,589	83.692	61.842
1500	29.325	33,516	85.711	63.367
1600	29.422	36,454	87.607	64.823
1700	29.521	39,401	89.394	66.217
1800	29.633	42,358	91.084	67.552
1900	29.773	45,328	92.690	68.833
2000	29.957	48,314	94.221	70.064
2100	30.203	51,322	95.689	71.250
2200	30.528	54,358	97.101	72.393
2300	30.954	57,431	98.467	73.497
2400	31.501	60,552	99.795	74.565
2500	32.191	63,736	101.095	75.601
2600	33.050	66,996	102.374	76.606
2700	34.103	70,522	103.640	77.584
2800	35.376	73,824	104.902	78.537
2900	36.898	77,436	106.170	79.468
3000	38.698	81,213	107.450	80.379
3100	40.808	85,186	108.753	81.273
3200	43.260	89,386	110.086	82.153
3293	45.876	93,528	111.362	82.960
3293	42.182	127,411	121.651	82.960
3300	42.182	127,707	121.741	83.042
3400	42.182	131,925	123.000	84.198
3500	42.182	136,143	124.223	85.325
3600	42.182	140,361	125.411	86.421
3700	42.182	144,580	126.567	87.491
3800	42.182	148,798	127.692	88.534
3900	42.182	153,016	128.787	89.552
4000	42.182	157,234	129.855	90.547
4100	42.182	161,452	130.897	91.518
4200	42.182	165,671	131.913	92.468
4300	42.182	169,889	132.906	93.397
4400	42.182	174,107	133.876	94.306
4500	42.182	178,325	134.824	95.196

Table 16 continued

Temperature K	$C°_p$ J/(mol K)	$H°_T - H°_{298.15}$ J/mol	$S°_T$ J/(mol K)	$-(G°_T - H°_{298.15})/T$ J/(mol K)
4600	42.182	182,543	135.751	96.067
4700	42.182	186,762	136.658	96.921
4800	42.182	190,980	137.546	97.759
4900	42.182	195,198	138.416	98.579
5000	42.182	199,416	139.268	99.385
5100	42.182	203,634	140.103	100.175
5200	42.182	207,823	140.922	100.951
5300	42.182	212,071	141.726	101.713
5400	42.182	216,289	142.514	102.461
5500	42.182	220,507	143.288	103.196
5600	42.182	224,725	144.048	103.919
5700	42.182	228,944	144.795	104.629
5800	42.182	233,162	145.529	105.328

improvement in the rapid pulse heating technique and show a relatively satisfactory agreement with the selected value. Thermodynamic values derived from Eq 7 are also given in Table 16.

4 Gas Phase

4.1 Thermodynamic Properties of the Gas Phase

Selected values are based on the 488 energy levels from the sources given in Table 4 with thermodynamic properties being calculated using the method of Kolsky et al.[56] and the 2014 Fundamental Constants (Mohr et al.[57,58]). Derived thermodynamic values are given in Table 17. Not included in Table 4 are eighteen levels given by Moore[59] which were shown to be spurious by Mocnik et al.[60] and a further five levels which were shown to be spurious by Jaritz et al.[61] It is noted that thermodynamic properties of the gas phase selected by both Gurvich et al.[25] and JANAF (Chase[26]) show differences from the presently accepted values which may be due to the fact that they selected only the energy levels of Moore[57] whereas the present evaluation included a further 228 levels.

4.2 Enthalpy of Sublimation

For values given in the form of the Clausius–Clapeyron equation a “pseudo” Third Law value was calculated by evaluating the enthalpy of sublimation at the temperature extremes and then averaging. Because of a general lack of detail as to what temperature scales were used and problems associated with the exact measurement of temperature then no attempt was made to correct vapor pressure measurements to ITS-90 from what would have been contemporary scales. However, the temperature values of Langmuir and Malter[62] and Fiske[63] were corrected from laboratory scales to IPTS-1948 using the method of Szwarc et al.[64] Values are summarised in Table 5. The selected value of 781 ± 4 kJ/mol is based on the close agreement between the values of Edwards et al.,[65] Gorbatyi and Shuppe[66] and the effusion measurements of Golubtsov and Nesmeyanov[67] with the selected accuracy taking into account the evaporation measurements of Golubtsov and

Table 17 Thermodynamic properties of the gas phase

Temperature K	C°_p J/(mol K)	$H^\circ_T - H^\circ_{298.15}$ J/mol	S°_T J/(mol K)	$-(G^\circ_T - H^\circ_{298.15})/T$ J/(mol K)
298.15	20.858	0	185.220	185.220
300	20.862	38.6	185.349	185.221
400	21.260	2141	191.395	186.042
500	22.082	4305	196.220	187.610
600	23.183	6567	200.341	189.396
700	24.381	8945	204.004	191.226
800	25.562	11,442	207.337	193.034
900	26.676	14,055	210.413	194.797
1000	27.701	16,775	213.278	196.503
1100	28.633	19,592	215.962	198.151
1200	29.468	22,498	218.490	199.742
1300	30.209	25,483	220.879	201.277
1400	30.858	28,537	223.142	202.759
1500	31.423	31,651	225.291	204.190
1600	31.912	34,819	227.335	205.573
1700	32.337	38,032	229.282	206.911
1800	32.708	41,284	231.141	208.206
1900	33.034	44,572	232.919	209.460
2000	33.325	47,890	234.621	210.676
2100	33.589	51,236	236.253	211.855
2200	33.830	54,607	237.821	213.000
2300	34.055	58,001	239.330	214.112
2400	34.268	61,418	240.784	215.193
2500	34.471	64,855	242.187	216.245
2600	34.668	68,312	243.543	217.269
2700	34.860	71,788	244.855	218.267
2800	35.049	75,283	246.126	219.239
2900	35.237	78,798	247.359	220.188
3000	35.425	82,331	248.557	221.113
3100	35.614	85,883	249.722	222.018
3200	35.807	89,454	250.855	222.901
3293	35.989	92,792	251.884	223.705
3300	36.003	93,044	251.960	223.765
3400	36.203	96,654	253.038	224.610
3500	36.409	100,285	254.090	225.438
3600	36.622	103,937	255.119	226.248
3700	36.840	107,610	256.125	227.042
3800	37.065	111,305	257.111	227.820
3900	37.297	115,023	258.077	228.584
4000	37.537	118,765	259.024	229.333
4100	37.782	122,530	259.954	230.068
4200	38.035	126,321	260.867	230.791
4300	38.293	130,138	261.765	231.501
4400	38.557	133,980	262.649	232.199
4500	38.826	137,849	263.518	232.885
4600	39.099	141,745	264.374	233.560
4700	39.376	145,669	265.218	234.225

Table 17 continued

Temperature K	C°_p J/(mol K)	$H^\circ_T - H^\circ_{298.15}$ J/mol	S°_T J/(mol K)	$-(G^\circ_T - H^\circ_{298.15})/T$ J/(mol K)
4800	39.656	149,621	266.050	234.879
4900	39.937	153,600	266.871	235.524
5000	40.219	157,608	267.680	236.159
5100	40.502	161,644	268.480	236.785
5200	40.783	165,709	269.269	237.402
5300	41.063	169,801	270.048	238.010
5400	41.341	173,921	270.819	238.611
5500	41.615	178,069	271.580	239.203
5600	41.885	182,244	272.332	239.788
5700	42.150	186,446	273.076	240.366
5800	42.409	190,674	273.811	240.936

$H^\circ_{298.15\text{ K}} - H^\circ_{0\text{ K}}$ 6199.6 J/mol

Nesmeyanov[67] and the measurements of Langmuir and Malter,[62] although the latter showed a marked trend with temperature.

5 Vapor Pressure

The vapor pressure equations for the solid as given in Table 6 was evaluated from free energy functions for the solid and the gas at 50 K intervals from 1800 K to 3250 K and the melting point and for the liquid at 50 K intervals from 3300 to 5800 K and the melting point and were fitted to the following equation:

$$\ln(p, \text{bar}) = A + B\ln(T) + C/T + D\,T + E\,T^2$$

6 Comments on Previous Reviews of the Thermodynamic Properties

Gurvich et al.[25] also appeared to give a preference towards the high temperature solid enthalpy measurements, and the selected values agree fairly satisfactory with the presently accepted values showing a maximum specific heat difference of 2.7% higher at 2800 K. However, JANAF (Chase[26]) gave preference to the high temperature specific heat values of Cezairliyan et al.[43] and as a result selected enthalpy values are on average 1630 J/mol higher than the smoothed experimental values above 3000 K. In the high temperature region, Maglić[27] selected mainly the specific heat measurements of Lehman[40] and Taylor and Finch[41,42] which were rejected here for reasons given in Part 2.3. As a result, selected specific heat values trend from 0.9% low initially to 3.3% low at 1000 K to 16.8% high at 3200 K. Integration of the specific heat equation given by Maglić[27] led to marked enthalpy differences compared to selected values trending 1950 J/mol to 3070 J/mol higher in the narrow range 3000 K to 3200 K. The review by Bodryakov[28] superseded an earlier review by the same author.[68] In the high temperature region, Bodryakov[28] selected specific heat values which differed sinusoidally from the values selected here trending to 2.2% low at 1000 K to 5.8% high at 2400 K to 1.8% low at 3250 K. Although the specific heat curve selected by Bodryakov behaved more naturally than the presently selected curve, integration leads to enthalpy values which exceed smoothed experimental values by an average of 1290 J/mol at 3000 K and above. On these grounds, the selected values of JANAF (Chase[26]), Maglić[27] and Bodryakov[28] are considered as not being representative of the actual high temperature experimental enthalpy values.

7 Summary of Representative Equations

High temperature specific heat equations are given in Table 7, free energy equations are given in Table 8 and transition values involved with the free energy equations in Table 9.

Table 18 Vapor Pressure

T K	p bar	ΔG_T J/mol	ΔH_T J/mol	p bar	T K
298.15	4.77×10^{-130}	738,166	781,000	10^{-15}	1826
300	3.33×10^{-129}	737,901	780,992	10^{-14}	1911
400	3.23×10^{-95}	723,606	780,532	10^{-13}	2005
500	7.73×10^{-75}	709,428	780,075	10^{-12}	2109
600	2.93×10^{-61}	695,338	779,662	10^{-11}	2224
700	1.45×10^{-51}	681,312	779,318	10^{-10}	2352
800	2.68×10^{-44}	667,331	779,050	10^{-9}	2496
900	1.20×10^{-38}	653,378	778,860	10^{-8}	2658
1000	3.98×10^{-34}	639,442	778,746	10^{-7}	2843
1100	1.98×10^{-30}	625,515	778,702	10^{-6}	3055
1200	2.39×10^{-27}	611,588	778,725	10^{-5}	3303
1300	9.69×10^{-25}	597,657	778,809	10^{-4}	3609
1400	2.67×10^{-22}	583,717	778,948	10^{-3}	3979
1500	1.44×10^{-20}	569,766	779,135	10^{-2}	4435
1600	7.17×10^{-19}	555,801	779,365	10^{-1}	5010
1700	2.25×10^{-17}	541,820	779,631	1	5756.93
1800	4.82×10^{-16}	527,823	779,926	NBP	5761.84
1900	7.49×10^{-15}	513,808	780,244		
2000	8.86×10^{-14}	499,777	780,576		
2100	8.29×10^{-13}	485,729	780,914		
2200	6.33×10^{-12}	471,665	781,249		
2300	4.06×10^{-11}	457,585	781,571		
2400	2.23×10^{-10}	443,492	781,865		
2500	1.07×10^{-9}	429,388	782,119		
2600	4.54×10^{-9}	415,275	782,315		
2700	1.74×10^{-8}	401,155	782,266		
2800	6.02×10^{-8}	387,033	782,459		
2900	1.92×10^{-7}	372,912	782,362		
3000	5.66×10^{-7}	358,797	782,118		
3100	1.56×10^{-6}	344,692	781,697		
3200	4.01×10^{-6}	330,605	781,068		
3293	9.19×10^{-6}	317,524	780,264		
3293	9.19×10^{-6}	317,524	746,381		
3300	9.74×10^{-6}	316,613	746,338		
3400	2.17×10^{-5}	303,600	745,730		
3500	4.60×10^{-5}	290,605	745,142		
3600	9.37×10^{-5}	277,626	744,575		
3700	1.84×10^{-4}	264,663	744,030		
3800	3.47×10^{-4}	251,714	743,507		
3900	6.34×10^{-4}	238,779	743,007		
4000	1.12×10^{-3}	225,856	742,531		
4100	1.94×10^{-3}	212,946	742,078		
4200	3.25×10^{-3}	200,044	741,651		
4300	5.33×10^{-3}	187,154	741,249		
4400	8.53×10^{-3}	174,272	740,873		
4500	1.34×10^{-2}	161,399	740,524		
4600	2.06×10^{-2}	148,533	740,202		

NBP = Normal boiling point at 1 atmosphere pressure (1.01325 bar)

Enthalpy of sublimation at 0 K: ΔH°_0 780,484 J/mol

Table 18 continued

T K	p bar	ΔG_T J/mol	ΔH_T J/mol	p bar	T K
4700	3.11×10^{-2}	135,674	739,908		
4800	4.61×10^{-2}	122,821	739,641		
4900	6.73×10^{-2}	109,972	739,403		
5000	9.67×10^{-2}	97,129	739,192		
5100	0.137	84,241	739,010		
5200	0.192	71,454	738,856		
5300	0.264	58,621	738,730		
5400	0.361	45,789	738,633		
5600	0.649	20,131	738,519		
5700	0.857	7303	738,502		
5756.93	1.000	0	738,505		
5800	1.121	− 5525	738,512		

8 Deviations from the Selected Values

Deviations of superconducting specific heat values are given in Table 10 and low temperature normal state specific heat in Table 11. Deviations of high temperature enthalpy and specific heat values in the solid range are given in Tables 12 and 13 respectively.

9 Thermodynamic Tables

Low temperature thermodynamic properties of the solid are given in Tables 14 and 15 and of the high temperature thermodynamic properties of the condensed phases in Table 16. Thermodynamic properties of the gas are given in Table 17 whilst the vapour pressure summary is given in Table 18.

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