Heat Capacity of Reference Materials: Cu and W

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The CODATA Task Group on Thermophysical Properties is preparing a set of recommended values for the heat capacity, thermal expansion, and transport properties of key solids which are used in calibrating or checking measuring equipment. The present paper surveys selected data on heat capacity at constant pressure C_p of copper from 1 to 1300 K and tungsten from 1 to 3400 K. Selected values are tabulated for C_p and also for heat capacity at constant volume C_v. Interpolating functions are given for C_p.

Key words: copper; critically evaluated data; heat capacity at constant pressure; heat capacity at constant volume; reference materials; tungsten.

1. Introduction

The Task Group on Thermophysical Properties, established by the Committee on Data for Science and Technology (CODATA) is preparing recommended values for some properties of a few key materials used widely for calibration or reference.[1,2] They have asked us to evaluate the heat capacity of copper and tungsten. The criteria for selecting data and format of their presentation largely follow previous reports arising from this Task Group on recommended values of expansivity of copper,[3] tungsten, and alumina.[4]

The heat capacity per mole at constant pressure is denoted by C_p and at constant volume by C_v. The quantity which is usually measured is either $C_p = (\mathrm{d}H/\mathrm{d}T)_p$, where H is the enthalpy, or the total change in enthalpy between temperature T and some reference temperature such as 273.15 K or 298.15 K. However $C_v = (\mathrm{d}U/\mathrm{d}T)_v$ (where U is the internal energy) is of theoretical importance. It may be calculated from C_p using

$$C_p = C_v(1 + \beta\gamma T),$$

where β is the volume expansion coefficient, B_T is the isothermal bulk modulus, and V is the molar volume. γ is the thermal Grüneisen parameter given by

$$\gamma = \beta B_s V/C_p \equiv \beta B_T V/C_v.$$

All these quantities are, in general, functions of temperature. Note that a similar thermodynamic relation enables us to calculate B_T from the adiabatic modulus $B_s = B_T(1 + \beta\gamma T)$.

The internal energy of a solid, and hence C_v, may contain contributions from magnetic interactions, free electrons, rotational states, phase changes, etc., in addition to the vibrations of the crystal lattice. For the reference solids discussed here, the vibrational energy is the major contribution above liquid-helium temperatures. The electronic term C_e is significant at liquid-helium temperatures because it is proportional to T whereas the lattice term C_l changes more rapidly as T^3. For $T \ll \theta_D$ (the Debye characteristic temperature),

$$C_l = BT^3 + CT^5 + DT^7 + \cdots$$
$$= \frac{12\pi^4}{5}\frac{NnkT^3}{\theta_0^3} \text{ as } T \to 0,$$
$$\approx 1944 \cdot n \cdot (T/\theta_0)^3$$

per mole of formula units,

where N = Avogadro's number, k = Boltzmann's constant, n = number of atoms per formula unit, and θ_0 is the value of the Debye temperature as $T \to 0$. Unlike transport properties, the heat capacity is not controlled in the first order by impurities. Traces of impurity at the <0.1 mol % level are usually insignificant in metals, unless the impurities are magnetic and the temperature is below 10 or 20 K, where C_l is small (e.g., see Fig. 1).

Our sources of input data are from the compilations of Touloukian and Buyco,[5] Hultgren *et al.*,[6] Furukawa *et al*,[7] and Reilly and Furukawa,[8] and some later references from Chemical Abstracts. From these we selected primary data sources which satisfied the criterion that random errors were generally less than 2%.

Systematic errors are more difficult to distinguish. We have generally excluded data which depart significantly from the mean in the intermediate temperature range of 300–1000 K where many "experienced" observers are in good agreement. The word experienced is applied particularly to those in national laboratories where there is proven competence in temperature measurement. Note also that at both low and high temperatures, theoretical considerations dictate a form of $C_p(T)$, serious departures from which lead us to reject data. A compilation of this type must be subjective as the selection and treatment reflect the experience—and bias—of the compilers. We reject a treatment which merely accepts and averages all available data. Table 1 lists for copper those primary sources for temperatures above 25 K, and Table 5 gives all primary sources for tungsten. For primary sources on copper below 25 K, we refer to the many papers on the 1965 Calorimetry Conference Copper discussed below.

The selected authors have generally reported their data for C_p in smoothed form as a table and/or a polynomial, but some give only a polynomial for the enthalpy $H(T)$ which we have differentiated to produce C_p values at selected tempera-

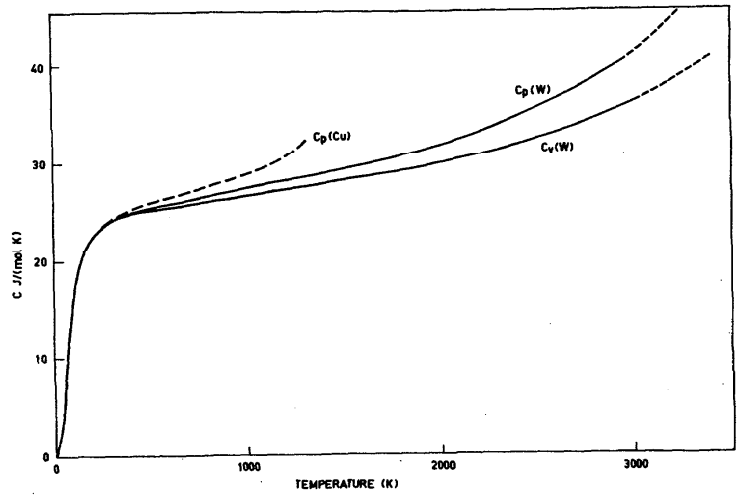

FIG. 1. The heat capacity of copper and tungsten.

TABLE 1. Selected literature data on copper (above 25 K)

Author (s)	Year	Range (K)	Comments
Bronson *et al.* (Ref. 17)	1933	273–773	Commerically pure cold-rolled Cu
Brooks *et al.* (Ref. 18)	1968	313–1173	Electrolytic Cu < 0.001% impurity
Klinkhardt (Ref. 19)	1927	373–1073	0.1% Ni: vacuum melted Cu
Kraftmakher (Ref. 20)	1967	500–1250	No details
Lucks and Deem (Ref. 21)	1956	273–1338	Electrolytic Cu
Martin (Ref. 22)	1960	20–300	99.999% ASARCO Cu
Pawel and Stansbury (Ref. 23)	1965	293–873	99.99% electrolytic Cu
Robie *et al.* (Ref. 24)	1976	15–375	1965 Calorimetry Conference Copper Sample

TABLE 2. Heat capacity of copper (J $mol^{-1} K^{-1}$) (m.p. 1356 K)

T (K)	C_p	C_v	T (K)	C_p	C_v
1	0.000 743		100	16.01	15.91
2	0.001 77		120	18.25	18.11
3	0.003 37		140	19.87	19.65
4	0.005 82		160	21.05	20.78
5	0.009 43		180	21.94	21.59
6	0.014 5		200	22.63	22.23
7	0.021 3		220	23.17	22.72
8	0.030 1		240	23.60	23.07
9	0.041 4		260	23.94	23.36
10	0.055 5		280	24.22	23.58
12	0.093 6		300	24.45	23.74
14	0.149		350	24.88	24.02
16	0.225		400	25.25	24.23
18	0.328		450	25.61	24.44
20	0.462		500	25.91	24.58
25	0.963		550	26.21	24.70
30	1.693		600	26.48	24.79
35	2.64		650	26.73	24.87
40	3.74	3.74	700	26.99	24.94
50	6.15	6.14	800	27.48	25.03
60	8.595	8.58	900	28.04	25.15
70	10.86	10.83	1000	28.66	25.30
80	12.85	12.80	1100	29.48	25.57
90	14.56	14.49	1200	30.53	25.98
			1250	31.12	26.22
			1300	32.16	26.80

tures. We have then averaged the data to produce a table of C_p and C_v and where possible an algebraic expression for recommended values of C_p. Procedures differ for different materials and temperature ranges as discussed below.

Values of C_p and C_v are given in Table 2 for copper in the range 1–1300 K and in Table 4 for tungsten in the range 1–3400 K.

2. Copper

2.1. 0.3–25 K

The 1965 Calorimetry Conference Copper Standard is well established as a reference material for evaluating the performance of calorimeters below 25 K. The Copper Standard was vacuum cast at the Argonne National Laboratory from ASARCO 99.999 + % copper.[9] Measurements between 1 and 25 K by Osborne *et al.*[9] and Ahlers[10] on this copper and by Martin[11] on other vacuum-cast ASARCO 99.999 + % copper, together with other data from the literature, were used by Osborne *et al.*[9] to produce the polynomial reference equation:

$$C_p = \sum_{i=1}^{6} A_i T^{2i-1} \text{ mJ/mol K}, \quad (1)$$

where (1 mol = 63.54 g)

$$A_1 = 6.9434\times10^{-1}, \quad A_4 = 9.4786\times10^{-8},$$
$$A_2 = 4.7548\times10^{-2}, \quad A_5 = -1.3639\times10^{-10},$$
$$A_3 = 1.6314\times10^{-6}, \quad A_6 = 5.3898\times10^{-14}.$$

This equation which gives values in Table 2 ($T<25$/K), fits the input data with standard deviation of 0.40% and fits the selected values given by Furukawa *et al.*[7] to ± 0.1%. Boerstoel *et al.*[12] and Hurley and Gerstein[13] examined later results from a variety of investigations in the temperature range 1–30 K and found that deviations from this copper reference equation rarely exceeded 0.3%.

Holste *et al.*[14] analyzed heat capacity data for copper from 1 to 30 K using five different experimental temperature scales and concluded that those scales based on a paramagnetic-salt thermometer led to maximum differences in C_p of 0.3%. They suggested that major differences in heat capacity data for copper between 1 and 4 K are probably due to differences in samples or techniques. Martin[15] has observed that the specific heat of copper in the "as received" condition may be higher than that measured in samples that have been vacuum annealed or vacuum cast, due to the presence of a dissolved gas.

Below 25 K copper is the best established material for testing calorimeters, with the C_p values those given by the copper reference equation. However a lower limit of 0.3 K should be set for the use of copper as a calorimetric standard because there is evidence of a heat capacity anomaly below this temperature.[16]

2.2. 25–300 K

Furukawa *et al.*[7] critically evaluated copper data up to 300 K, and produced a table of selected values (but not an interpolating function) for C_p and other thermodynamic functions. They selected sets of data which were not scattered by more than 2.5% and averaged them. Measurements by Martin[22] on 99.999% pure copper, and more recently by Robie *et al.*[24] on the 1965 Calorimetry Conference Copper in the range 20–300 K were within ± 1% of the Furukawa values. We have accepted these values of Furukawa *et al.* (reproduced in Table 2) and have least-squares fitted them (rms deviation of 0.07%) to an eighth-order interpolating polynomial

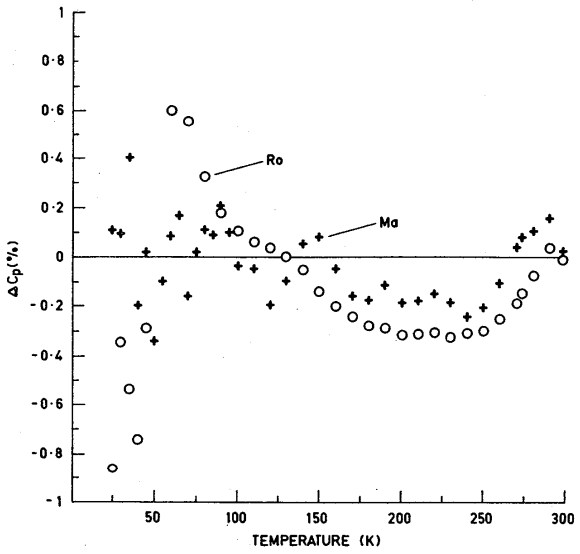

FIG. 2. Heat capacity of copper. Deviations of Robie *et al.* (Ref. 24) (○), and Martin (Ref. 22) (+) from selected values (Ref. 7).

$$C_p = \sum_{n=0}^{8} a_n (T/100)^n \text{ J/mol K}. \quad (2)$$

The coefficients are

$$a_0 = 4.892\,87, \quad a_5 = -132.542\,5,$$
$$a_1 = -57.517\,01, \quad a_6 = 38.173\,99,$$
$$a_2 = 238.203\,9, \quad a_7 = -6.079\,62,$$
$$a_3 = -345.428\,3, \quad a_8 = 0.411\,868\,7.$$
$$a_4 = 275.897\,5,$$

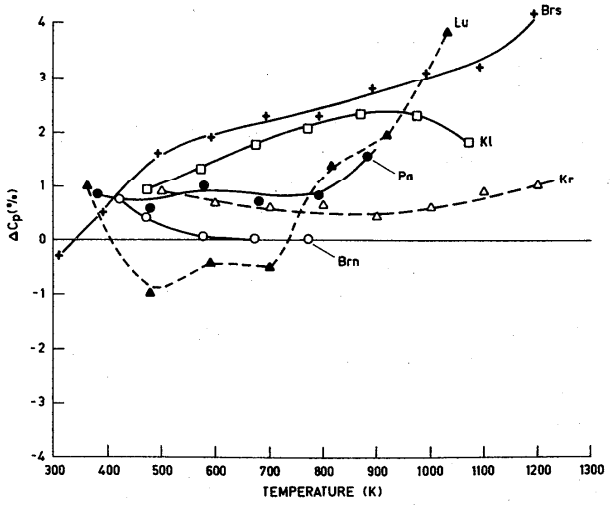

FIG. 3. Heat capacity of copper. Deviations of various C_p data (excluding those derived from enthalpy) from Hultgren *et al.* (Ref. 6) from 300–1300 K, Kl: Ref. 19, Kr: Ref. 35, Brs: Ref. 18, Lu: Ref. 21, Brn: Ref. 17, Pa: Ref. 23.

TABLE 3. Spline coefficients of copper for C_p (J mol^{-1} K^{-1}) as a function of $T/100$[a]

N	$T_{min}/100$	$T_{max}/100$	a_0	a_1	a_2	a_3
1	0.2500	0.2900	0.962 97	12.4259	39.904 50	64.762 60
2	0.2900	0.4473	1.528	15.9290	47.676 00	− 89.748 40
3	0.4473	0.6920	4.864	24.2660	5.323 70	− 29.051 00
4	0.6920	1.3946	10.695	21.6530	− 16.003 00	5.242 50
5	1.3946	2.0000	19.827	6.9300	− 4.952 40	1.873 60
6	2.0000	3.3000	22.623	2.9936	− 1.549 60	0.372 40
7	3.3000	12.3750	24.714	0.8526	− 0.097 37	0.008 73
8	12.3750	13.0000	30.960	1.2433	0.140 40	1.508 00

[a] $C_p(T/100) = a_0 + a_1 t + a_2 t^2 + a_3 t^3$ for the range $T_{min}/100 \leqslant T/100 < T_{max}/100$ and $t = (T - T_{min})/100$.

We estimate the error to be $<1\%$ in recommended values below 100 K and approximately 0.3% between 100 and 300 K. Figure 2 shows the deviations from the interpolating polynomial of the data of Martin[22] and Robie *et al.*[24] Furukawa *et al.*[7] give a deviation plot from 30 to 300 K for another 18 sets of data which are generally within $\pm 2\%$ of the selected values.

2.3. 300–1300 K

The recommended values in Table 2 are those produced by Hultgren *et al.*[6] from selected enthalpy and heat capacity data published in the literature up to 1968. Figure 3 shows the deviation of some selected sets of C_p data from those recommended. The systematic positive deviation indicates that the enthalpy data used by Hultgren lead to smaller C_p values and suggest that the recommended values could be in error by as much as 2% above 800 K.

2.4. Alternative Interpolation by Cubic Splines

An alternative method of interpolating for the whole range from 25 to 1300 K is by the method of least squares using cubic splines of the form

$$a_0 + a_1 t + a_2 t^2 + a_3 t^3$$

(see Table 3). The input data for the fit are the selected values from Furukawa *et al.*[7] and Hultgren *et al.*[6] (see Table 2). The cubic spline was constrained to have the functional value, and the value of the first derivative of the copper reference Eq. (1) at 25 K to ensure proper matching. Table 3 lists the coefficients and knots of the fitted splines, which fit the selected values within $\pm 0.08\%$ with a mean deviation of 0.007%.

We have included both polynomial and spline interpolating functions to give readers the flexibility of using either the simpler polynomial expression from 25–300 K or the more complex spline functions for the whole range 25–1300 K.

2.5. Values of C_v

To calculate the values of C_v, we first calculated $\gamma = 3\alpha B_s V/C_p$ using recommended values of linear expansivity, α,[3] and of bulk modulus, B_s[25]; the latter only extend up to 800 K and were extrapolated linearly to 1300 K with a probable uncertainty of 2%. The molar volume $V = 7.09$ cm^3 at 300 K, increasing to 7.53 cm^3 at 1300 K. The γ values increase slightly from 1.98 at 150 K to 2.02 at 1200 K. Errors in $\gamma\beta T$ should be $\leqslant 1\%$ at 1000 K and therefore errors in $1 + \beta\gamma T = C_p/C_v$ should not exceed 0.1% at 1000 K rising to 0.4% at 1300 K.

3. Tungsten

3.1. 1–25 K

The values that we recommended for C_p in Table 4 are from Triplett *et al.*,[26] who fitted their results from 0.35 to 25 K by the expression

$$C_p = 1.008T + 0.0346T^3 + 2.84\times10^{-8}T^7 \text{ mJ/mol K.}$$

The analysis by Reilly and Furukawa[8] and review by Phillips[27] indicate differences among experimental values which increase rapidly below 25 K, reaching more than 10% at liquid-helium temperatures. We estimate the recommended values from Triplett *et al.* should be representative of pure tungsten within 5% at 10 K, and 2% at 20 K, but do not suggest that tungsten is a well established reference material in this low temperature region. Copper is to be preferred.

3.2. 25–300 K

The selected values (Table 4) are from an analysis by Reilly and Furukawa[8] and differ by less than 1% at most temperatures from those of Hultgren *et al.*[6] Both are based largely on primary data of Clusius and Franzosini[28] and DeSorbo [29] (see Fig. 4).

We have fitted the values of Reilly and Furukawa by the method of least squares to an interpolating polynomial of the same form used for copper, Eq. (2), over the 25–300 K temperature range. The coefficients are

$a_0 =$	7.828 80,	$a_5 =$	− 185.286 7,
$a_1 =$	− 83.953 18,	$a_6 =$	54.498 18,
$a_2 =$	317.322 1,	$a_7 =$	− 8.853 86,
$a_3 =$	− 463.671 5,	$a_8 =$	0.610 992,
$a_4 =$	377.553 1,	(units J/mol K).	

The polynomial fits the recommended values above 60 K with an inaccuracy of $<0.1\%$, $\sim 0.5\%$ between 30–60 K, and 1% between 25–30 K. The rms deviation over the whole range is 0.25%.

TABLE 4. Heat capacity of tungsten (J mol^{-1} K^{-1}) (m.p. 3680 K)

T (K)	C_p	C_v	T (K)	C_p	C_v
1	0.001 04		220	23.04	22.93
2	0.002 29		240	23.47	23.35
3	0.003 96		260	23.81	23.68
4	0.006 25				
5	0.009 37		280	24.10	23.96
6	0.013 5		300	24.35	24.20
7	0.018 9		350	24.69	24.52
8	0.025 8		400	24.96	24.74
9	0.034 4		500	25.44	25.16
10	0.045 0		600	25.87	25.51
12	0.072 9		700	26.27	25.86
14	0.112		800	26.64	26.14
16	0.165		900	27.01	26.45
18	0.237		1000	27.36	26.70
20	0.333		1100	27.72	26.99
25	0.73		1200	28.08	27.25
30	1.35		1300	28.45	27.54
35	2.22		1400	28.84	27.82
40	3.30		1500	29.24	28.12
45	4.53		1600	29.67	28.42
50	5.82		1700	30.13	28.72
60	8.39	8.39	1800	30.63	29.06
70	10.74	10.73	2000	31.74	29.80
80	12.81	12.80	2200	33.04	30.71
90	14.57	14.56	2400	34.57	31.79
100	16.04	16.02	2600	36.36	33.02
120	18.28	18.24	2800	38.44	34.41
140	19.87	19.82	3000	40.86	35.91
160	21.01	20.95	3200	(44.3)	(37.8)
180	21.86	21.78	3400	(48.3)	(40.0)
200	22.51	22.41			

TABLE 5. Selected literature data on tungsten

Author (s)	Year	Range	Form	Representation
Bronson *et al.* (Ref. 17)	1933	250–770 K	Sintered rod	H, polynomial
Cezairliyan & McLure (Ref. 30)	1971	2000–3600	Tube	C_p, polynomial
Clusius & Franzosini (Ref. 28)	1959	11–273	Sintered rod	C_p, polynomial
DeSorbo (Ref. 29)	1958	13–93 K	Powder	C_p, table
Ditmars (Ref. 31)	1979	273–1173	Crystal	H, polynomial
Ditmars (Ref. 31)	1979	273–1173	Sintered	H, polynomial
Hein & Flagella (Ref. 32)	1968	1200–3250	Sintered	H, polynomial
Jaeger & Rosenbohm (Ref. 33)	1930	600–900	...	C_p, table
Kirillin *et al.* (Ref. 34)	1963	600–3500 K	Sintered	H, polynomial
Kraftmakher (Ref. 35)	1973	1500–3600	Wire	C_p, table
Lowenthal (Ref. 36)	1963	1200–2400	Wire	C_p, table
Taylor (Ref. 37)	1979	1550–2400	Sintered	C_p, polynomial
Triplett *et al.* (Ref. 26)	1973	0.3–25	Polycrystal	C_p, polynomial
West & Ishihara (Ref. 38)	1968	1100–2500	99.95% and zone-refined rods	H, polynomial

3.3. 300–3500 K

Ten selected sets of data (Table 5) have been fitted by least squares (with $1/T^2$-weighting to prevent the rather scattered high temperature data from adversely affecting the fit at lower temperatures, see Fig. 5), to the polynomial

$$C_p = \sum_{n=-1}^{3} A_n t^n \text{ J/mol K} (t = T/1000),$$

over the range 300 to 3000 K:

$A_{-1} = -0.208\,69$, $A_2 = -1.999\,22$,
$A_0 = 23.703\,45$, $A_3 = 0.734\,168$,
$A_1 = 5.132\,062$, (and one mole = 183.85 g).

The individual deviations are shown in Fig. 5 and rms deviation is 1.1%. After initial trials, the data of Kirillin *et al.*[34] were excluded above 2500 K as their values were increasingly low compared with others.[30,32,35]

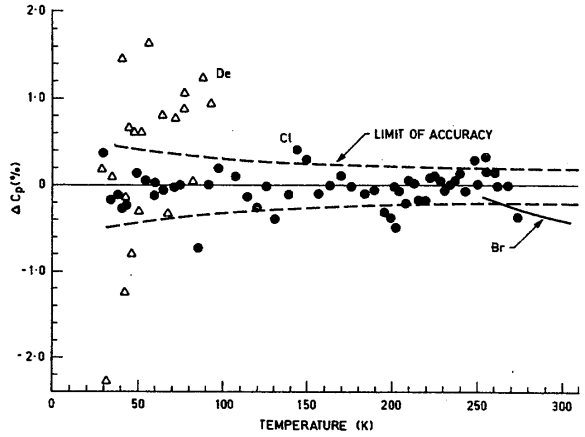

FIG. 4. Heat capacity of tungsten. Deviations of data of Clusius and Franzosini (Ref. 28) (Cl) and DeSorbo (Ref. 29) (De) and Bronson *et al.* (Ref. 17) (Br) from recommended values.

Values recommended by Hultgren *et al.* are not shown in Fig. 5 as they are much smaller than the fitted values above 2000 K, reaching 15% low at 3000 K. Below 1800 K they do not differ much from those of Jaeger and Rosenbohm[33] or Ditmars.[31]

We do not recommend values or attempt an algebraic fit above 3000 K because there are only two sets of data extending to 3500 K and these diverge, the difference reaching 10% at 3400 K. The two figures in brackets in the table are indicative values from Cezairliyan and McLure.[30]

3.4. General

The purity of the materials listed in Table 5 generally exceeded 99.9%. There is no evidence that the heat capacity of tungsten is affected significantly by trace impurities. Ditmars[31] did observe that C_p for his single crystal was 0.3% larger than for polycrystalline sintered rod from 273–1173 K, but this difference is less than the probable errors given in our table.

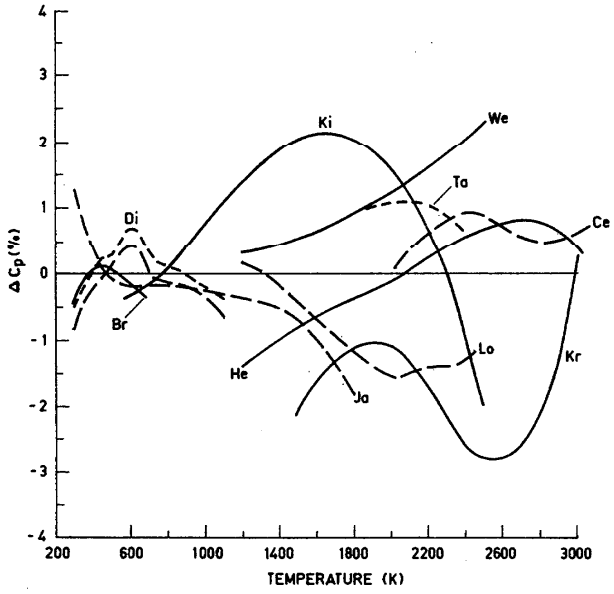

FIG. 5. Heat capacity of tungsten. Deviations from the recommended values of data taken from sources listed in Table 5. Di[31], Br[17], Ki[34], He[32], Ja[33], We[38], Lo[36], Ta[37], Ce[30], Kr[35].

3.5. Values of C_v

To calculate the C_v values, we first calculated $\gamma = 3\alpha B_s V/C_p$ using recommended values of linear expansivity α[4] and of bulk modulus B_s.[39] The elastic data extend up to 2100 K and were extrapolated linearly above that temperature with a probable uncertainty of 2%. Molar volume V is 9.55 cm^3 at 300 K, increasing to 10.03 cm^3 at 3000 K (using $\Delta V/V = 3\Delta l/l$ data from White and Roberts[4]). The γ values lie between 1.58 and 1.62 from 100 to 1600 K, increasing slowly to 1.73 at 3000 K. Errors in $\gamma\beta T$ should be $\leqslant 2\%$ below 2000 K and reach 4% at 3000 K. Hence errors in the correction factor $1+\beta\gamma T$ will be $\leqslant 0.1\%$ up to 2000 K, reaching 0.6% at 3000 K.

4. Acknowledgments

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