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Evaluation of the Thermodynamic Properties of Tungsten

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A new evaluation of the thermodynamic properties of tungsten has been made. A set of parameters describing the Gibbs energy of each individual phase as a function of temperature and pressure is given. The experimental information on the P, T phase diagram and the thermodynamic data are compared with calculations made using the presented set of parameters.

KEY WORDS: enthalpy; equation of state; heat capacity; high pressure; high temperature; thermodynamic properties, tungsten.

1. INTRODUCTION

The thermodynamic properties of tungsten have been the subject of many experimental studies. The available experimental data consist of measurements of enthalpies, heat capacities, volumes and compressibilities at atmospheric pressure, and the effect of pressure on the volume of the solid phase at room temperature. Some experimental information concerning the pressure–temperature phase diagram has also been published.

A reliable description of the thermodynamic properties of the elements is essential in phase diagram calculations. The purpose of the present work is to provide a complete description of the thermodynamic properties of tungsten including solid, liquid, and gaseous phases.

2. THERMODYNAMIC MODELS

A detailed description of the thermodynamic models used in the present evaluation has already been presented in a paper by Fernandez

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Guillermet [1]. A short review of the model and its application to tungsten is given here.

The model gives an analytical expression for the Gibbs energy derived from the temperature and pressure dependence of those derivatives of G_m which have been studied experimentally. The data available for the individual phases of tungsten consists of measurements of heat capacity (C_P), heat content ($H_T - H_{298.15}$), molar volume (V_m), and compressibility (K) at atmospheric pressure (P_0) and some measurements of V_m at higher pressures. An expression for $G_m(T, P)$ can be obtained by integrating the relations

$$\left(\frac{\partial^2 G_m}{\partial T^2}\right)_P = -\frac{C_P(T, P)}{T}, \qquad \left(\frac{\partial G_m}{\partial P}\right)_T = V_m(T, P) \tag{1}$$

where $C_P(T, P)$ and $V_m(T, P)$ are functions chosen to represent the experimental information. The analytical form of the functions $C_P(T, 0)$ and $V_m(T, P)$ adopted in this evaluation have been described previously by Fernandez Guillermet [1].

2.1. Solid and Liquid Phases

The contributions to the Gibbs energy discussed in Ref. 1 give, when combined, the following type of analytical expression for the Gibbs energy of solid and liquid tungsten as a function of temperature and pressure:

$$\begin{aligned} G_m(T, P) - H_m^{SER} &= a + bT + cT\ln(T) + dT^2 + eT^3 + fT^4 + gT^{-1} \\ &\quad + jT^{(i+1)} + \int_0^P V_m(T, P)\,dP \end{aligned} \tag{2}$$

where

$$\begin{aligned} \int_0^P V_m(T, P)\,dP &= \frac{A\exp(\alpha_0 T + 1/2\,\alpha_1 T^2)}{(K_0 + K_1 T + K_2 T^2)(n-1)} \\ &\quad \times [(1 + nP(K_0 + K_1 T + K_2 T^2)^{1-1/n} - 1] \end{aligned} \tag{3}$$

and A is a parameter of the dimension volume. H_m^{SER} is the molar enthalpy of the stable state (bcc tungsten) at 298.15 K and 10^5 Pa. The exponent i is equal to -10 for bcc W above T_m and equal to 6 for the liquid below T_m. j is equal to zero for bcc W below T_m and for liquid W above T_m. The remaining parameters are identical to those given in Eqs. (2) and (6)–(10) in Ref. 1.

2.2. Gas Phase

The heat capacity, enthalpy, entropy, and Gibbs energy of gaseous tungsten obtain by calculating partition functions from spectroscopic data and the enthalpy of formation obtained by a second- or third-law analysis of experimental vapor pressure data have been published in *JANAF* [2]. Rand [3] fitted these data to an expression of the type

$$C_P = -(c + 2dT + 6\,eT^2 + 2gT^{-2}) \tag{4}$$

using the number of temperature intervals required for a fit within 0.2%. These results were accepted in the present assessment.

The present work is based on the Gibbs energy. Such a description cannot reproduce the critical point where the differences between liquid and gaseous phase disappear. It is thus of limited value to try to extend the description of the gas up to pressures above the range of applicability of the ideal gas law,

$$V_{\mathrm{m}}(P, T) = \frac{RT}{P} \tag{5}$$

This expression was thus adopted for describing the molar volume of gaseous tungsten.

The contributions to the Gibbs energy in the gas phase give when combined, the following type of analytical expression for the Gibbs energy:

$$G_{\mathrm{m}}^{\mathrm{gas}}(T, P) - H_{\mathrm{m}}^{\mathrm{SER}} = a + bT + cT\ln(T) + dT^2 + eT^3 + gT^{-1} + RT\ln(P) \tag{6}$$

The values of the constants *a* and *b* were evaluated by use of the vapor pressure and entropy data given in *JANAF* [2].

3. SELECTED EXPERIMENTAL DATA

3.1. Thermochemical Data

3.1.1. bcc and Liquid Tungsten

The enthalpy and the heat capacity of tungsten (W) have been measured many times throughout the years. An assessment using reliable experimental data available on bcc W has recently been published by Gurvich et al. [4]. Values for the difference in enthalpy and entropy between 0 and 298.15 K and heat capacities between 200 and 2500 K were taken from the selected data by Gurvich et al.

$$\begin{aligned} H^0(298.15) - H^0(0) &= 4.97\ \mathrm{kJ \cdot mol^{-1}} \\ S^0(298.15) &= 32.64\ \mathrm{J \cdot mol^{-1} \cdot K^{-1}} \end{aligned} \tag{7}$$

The heat capacities and enthalpies selected by Gurvich et al. above 2500 K were disregarded due to the increasing descrepancy with the recent experimental C_P data published by Cezairliyan and McClure [5] and the recent enthalpy measurements by Arpaci and Frohberg [6]. The data from Refs. 5 and 6 were thus included in the selected set above 2500 K instead of the data used by Gurvich et al.

The temperature of melting, $T_m = 3695$ K, selected by Gurvich et al. was accepted. The ethalpy of melting selected by Gurvich et al. dates from an estimation by Wouch et al. [7] based on solidification and cooling curves of containerless melts. This value seems rather uncertain compared to the more recent measurements performed by Arpaci and Frohberg [6] using a levitation technique combined with a drop calorimeter. The experimental enthalpies of liquid and solid W published by Arpaci and Frohberg were accepted and included in the optimization.

3.1.2. *Estimation of the Stability of hcp and fcc Tungsten*

The lattice stability of hcp and fcc tungsten relative to that of bcc tungsten has been evaluated by Kaufman and Bernstein [8] from an estimation of the metastable melting point of hcp and fcc W and the entropy difference between fcc, hcp, and bcc W. In their evaluation, Kaufman and Bernstein used a different enthalpy of melting of W. In the present work we have $H^0(\mathrm{liq}) - H^0(\mathrm{bcc}) = 52300\ \mathrm{J \cdot mol^{-1}}$, evaluated from the data given by Arpaci and Frohberg [6], whereas Kaufman and Bernstein used $30543\ \mathrm{J \cdot mol^{-1}}$, based on an assumed entropy of fusion of $8.368\ \mathrm{J \cdot mol^{-1}}$. We thus have to reevaluate the lattice stability of hcp and fcc W in order to reproduce Kaufman and Bernstein's original assumptions.

Kaufman and Bernstein estimated the melting point of metastable hcp tungsten as 2650 K, by extrapolation of the solid/liquid phase boundaries in some binary systems containing W and elements with hcp structure, and the entropy difference between bcc and hcp W as $0\ \mathrm{J \cdot mol^{-1} \cdot K^{-1}}$, by considering the variation with group number known, from other elements. By combining Kaufman nd Bernstein's estimated melting point and entropy difference with the present description of liquid and bcc W, we arrive at $H^0(\mathrm{hcp}) - H^0(\mathrm{bcc}) = 14750\ \mathrm{J \cdot mol^{-1}}$, whereas Kaufman and Bernstein obtained $8368\ \mathrm{J \cdot mol^{-1}}$.

Kaufman and Bernstein estimated the entropy and enthalpy difference between fcc and hcp from a consideration of the fcc/liquid equilibria in a number of binary systems containing W and elements with fcc structure. Accepting their estimated melting point at 2230 K and entropy difference $S^0(\text{hcp}) - S^0(\text{fcc}) = 0.63\ \text{J} \cdot \text{mol}^{-1} \cdot \text{K}^{-1}$, we arrive at $H^0(\text{hcp}) - H^0(\text{fcc}) = -4550\ \text{J} \cdot \text{mol}^{-1}$, whereas Kaufman and Bernstein obtained $-2092\ \text{J} \cdot \text{mol}^{-1}$.

3.2. Experimental Data Concerning V_m

The molar volume and the thermal expansion of bcc W have been measured many times. A detailed compilation of experimental results performed by Touloukian et al. is available in the TPRC data series [9]. The experimental data by Kirby [10] and Petukhov and Chekhovskoi [11] were selected from this compilation and included in the optimization.

The effect of pressure on the molar volume of bcc W has been studied by Ming and Manghnani [12] between 1.3 and 9.4 GPa under static conditions at 293 K, using X-ray measurements. The temperature dependence of the compressibility at atmospheric pressure has been measured by Bernstein [13] between 280 and 1473 K and by Lowrie and Gonas [14] between 297 and 2073 K. The compressibility given by Lowrie and Gonas at 293 K is in excellent agreement with the compressibility evaluated from the data of Ming and Manghnani. The data of Lowrie and Gonas were thus considered more reliable and were included in the optimization.

4. RESULTS

The evaluation of the various parameters was made by means of a computer program for the optimization of thermodynamic parameters developed by Jansson [15, 16]. All the parameter values obtained in the present evaluation are summarized in Tables I–III. Table IV shows the calculated properties of the stable modification of tungsten at 1 atm. Comparisons with experimental data are given in Figs. 1 to 7.

Figure 1 shows the calculated heat capacity for tungsten in comparison with the experimental data of Schmidt et al. [17] and Cezairliyan and McClure [5] and the values recommended by Gurvich et al. [4]. The agreement with their values is excellent except for the points at very high temperatures. The reason for the rather low values selected by Gurvich et al. in this temperature range can be found in Fig. 2.

Figure 2 shows the calculated enthalpy in comparison with experimental data in the vicinity of the melting point. Gurvich et al. probably gave more attention to the high-temperature enthalpy measurements by

Table I. Summary of Parameters for Tungsten [All Data in SI Units; $R = 8.31448\ \mathrm{J \cdot mol^{-1} \cdot K^{-1}}$ (Proposed CODATA Value, 1985)][a]

Element, W	Stable element reference, bcc at 298.15 K and 10^5 Pa	Mass, 183.85	$H_{298} - H_0$, 4970	S_{298} 32.62

$298 < T < 3695$ K

$$G_{\mathrm{m}}^{\mathrm{bcc}} - H_{\mathrm{m}}^{\mathrm{SER}} = -7647.26 + 130.4\,T - 24.1\,T\ln(T) - 1.936 \times 10^{-3}\,T^2 + 2.07 \times 10^{-7}\,T^3 - 5.33 \times 10^{-11}\,T^4 + 44500\,T^{-1} + \int_0^P V_{\mathrm{m}}(T, P)\,dP \tag{8}$$

$3695 < T < 6000$ K

$$G_{\mathrm{m}}^{\mathrm{bcc}} - H_{\mathrm{m}}^{\mathrm{SER}} = -82{,}869.76 + 389.3623\,T - 54\,T\ln(T) + 1.921718 \times 10^{33}\,T^{-9} + \int_0^P V_m(T, P)\,dP$$

$298 < T < 3695$ K

$$G_{\mathrm{m}}^{\mathrm{liq.}} - H_{\mathrm{m}}^{\mathrm{SER}} = +44{,}513.324 + 116.29001\,T - 24.1\,T\ln(T) - 1.936 \times 10^{-3}\,T^2 + 2.07 \times 10^{-7}\,T^3 - 5.33 \times 10^{-11}\,T^4 + 44500\,T^{-1} - 2.713468 \times 10^{-24}\,T^7 + \int_0^P V_{\mathrm{m}}(T, P)\,dP \tag{9}$$

$3695 < T < 6000$ K

$$G_{\mathrm{m}}^{\mathrm{liq.}} - H_{\mathrm{m}}^{\mathrm{SER}} = -30{,}437 + 375.175\,T - 54\,T\ln(T) + \int_0^P V_{\mathrm{m}}(T, P)\,dP$$

$$G_{\mathrm{m}}^{\mathrm{fcc}} - G_{\mathrm{m}}^{\mathrm{bcc}} = +19{,}300 + 0.63\,T \tag{10}$$

$$G_{\mathrm{m}}^{\mathrm{hcp}} - G_{\mathrm{m}}^{\mathrm{bcc}} = +14{,}750 \tag{11}$$

$$G_{\mathrm{m}}^{\mathrm{gas}} - H_{\mathrm{m}}^{\mathrm{SER}} = a + bT + cT\ln(T) + dT^2 + eT^3 + gT^{-1} + RT\ln(P) \tag{6}$$

[a] Functions valid for $298.15 < T < 6000.00$ K unless other limits stated. The values of the constants are given in Table III.

Table II. Parameter Values Describing the Molar Volume of Each Individual Phase as a Function of T and P in Terms of Eq. (3) for Tungsten[a]

	A	α_0	α_1	K_0	K_1	K_2	n
bcc	9.5168×10^{-6}	9.386×10^{-6}	5.51×10^{-9}	3.1575×10^{-12}	1.6×10^{-16}	3.1×10^{-20}	4
Liq.	10.3×10^{-6}	9.386×10^{-6}	5.51×10^{-9}	3.1575×10^{-12}	1.6×10^{-16}	3.1×10^{-20}	4

[a] The molar volumes of fcc and hcp W are assumed to be the same as in bcc.

Table III. Functional Representation of the Thermodynamic Properties of Tungsten–Vapor in Terms of Eq. (6)

Temperature (K)	a	b	c	$d * 10^3$	$e * 10^7$	g
$298 < 600$	+848579.704	−196.512472	−10.25862	−8.767154	−40.04325	−163731.01
$600 < 1000$	+868664.325	−551.545553	+46.25286	−78.92865	+122.2551	−1491104.4
$1000 < 2000$	+763338.282	+565.444039	−116.1336	+32.71260	−22.09068	+11364727
$2000 < 3600$	+882843.778	−122.194186	−25.45370	+1.762248	−2.060976	−18534827
$3600 < 6000$	+986167.693	−508.439277	+22.31499	−8.031690	+1.694639	−60369890

Leibowitz et al. [18] than the corresponding C_P measurements by Cezairliyan and McClure [5]. The new measurements by Arpaci and Frohberg [6] support the data of Cezairliyan and McClure. The calculated enthalpy in bcc W at the melting point falls between the values reported by Leibowitz et al. and those reported by Arpaci and Frohberg, whereas the calculated enthalpy in liquid W is in excellent agreement with Arpaci and Frohberg's data. The enthalpy of melting calculated in this work is slightly larger than that suggested by Arpaci and Frohberg. The calculated values were accepted as a good compromise between the set of data shown in Fig. 1 and Fig. 2.

Table IV. Calculated Thermodynamic Properties of the Stable Modification of Tungsten at 1 atm

T (K)	$G - H^{\mathrm{SER}}$ ($\mathrm{kJ \cdot mol^{-1}}$)	$H - H_{298.15}$ ($\mathrm{kJ \cdot mol^{-1}}$)	$S - S_0$ ($\mathrm{J \cdot mol^{-1} \cdot K^{-1}}$)	C_P ($\mathrm{J \cdot mol^{-1} \cdot K^{-1}}$)
298.15	−9.7249	0.00001	32.618	24.160
400	−13.431	2.5036	39.835	24.935
500	−17.705	5.0239	45.457	25.449
600	−22.491	7.5903	50.135	25.867
700	−27.710	10.196	54.151	26.240
800	−33.305	12.838	57.678	26.591
900	−39.233	15.514	60.830	26.935
1000	−45.461	18.225	63.686	27.281
1100	−51.962	20.970	66.302	27.634
1200	−58.715	23.752	68.722	28.001
1300	−65.701	26.571	70.979	28.387
1400	−72.906	29.430	73.097	28.796
1500	−80.317	32.331	75.099	29.233
1600	−87.922	35.278	77.000	29.701
1700	−95.714	38.273	78.816	30.205
1800	−103.68	41.320	80.557	30.748

Table IV. *(Continued)*

T (K)	$G-H^{\mathrm{SER}}$ $(\mathrm{kJ\cdot mol^{-1}})$	$H-H_{298.15}$ $(\mathrm{kJ\cdot mol^{-1}})$	$S-S_0$ $(\mathrm{J\cdot mol^{-1}\cdot K^{-1}})$	C_P $(\mathrm{J\cdot mol^{-1}\cdot K^{-1}})$
1900	−111.82	44.424	82.235	31.336
2000	−120.13	47.589	83.858	31.971
2100	−128.59	50.820	85.435	32.657
2200	−137.21	54.122	86.971	33.399
2300	−145.99	57.501	88.473	34.201
2400	−154.91	60.964	89.947	35.065
2500	−163.97	64.517	91.397	35.997
2600	−173.19	68.166	92.828	37.000
2700	−182.54	71.919	94.244	38.077
2800	−192.03	75.784	95.650	39.233
2900	−201.67	79.769	97.048	40.472
3000	−211.44	83.881	98.442	41.797
3100	−221.36	88.131	99.835	43.213
3200	−231.41	92.517	101.23	44.722
3300	−241.60	97.079	102.63	46.329
3400	−251.94	101.80	104.04	48.038
3600	−273.03	111.77	106.89	51.777
3695	−283.24	116.77	108.26	53.708
3695	−283.24	169.09	122.42	54.000
3700	−283.86	169.36	122.49	54.000
3800	−296.18	174.76	123.93	54.000
3900	−309.65	180.16	125.34	54.000
4000	−321.25	185.56	126.70	54.000
4100	−333.99	190.96	128.04	54.000
4200	−346.86	196.36	129.34	54.000
4300	−359.85	201.76	130.61	54.000
4400	−372.98	207.16	131.85	54.000
4500	−386.22	212.56	133.06	54.000
4600	−399.59	217.96	134.25	54.000
4700	−413.07	223.36	135.41	54.000
4800	−426.67	228.76	136.55	54.000
4900	−440.38	234.16	137.66	54.000
5000	−454.20	239.56	138.75	54.000
5100	−468.13	244.96	139.82	54.000
5200	−482.17	250.36	140.87	54.000
5300	−496.31	255.76	141.90	54.000
5400	−510.55	261.16	142.91	54.000
5500	−524.89	266.56	143.90	54.000
5600	−539.33	271.96	144.87	54.000
5700	−553.86	277.36	145.83	54.000
5800	−568.49	282.76	146.77	54.000
5900	−583.21	288.16	147.69	54.000

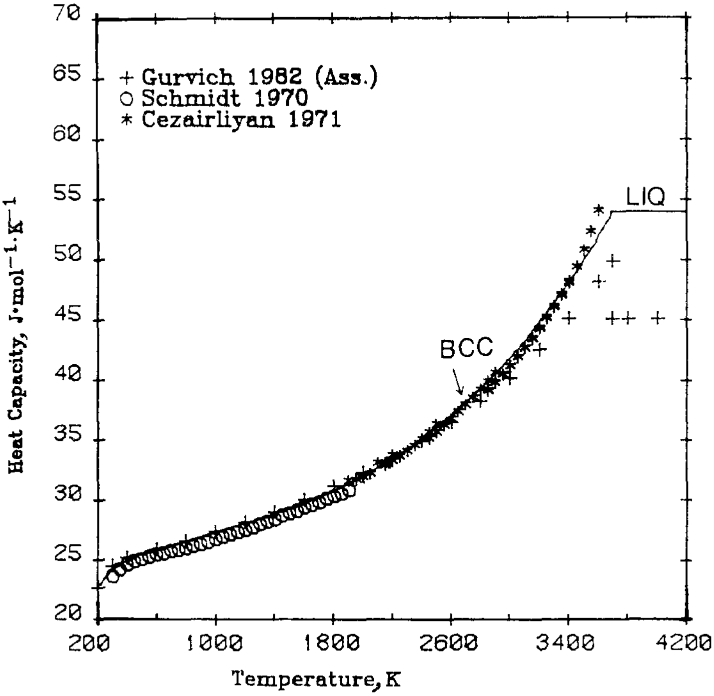

Fig. 1. Calculated heat capacity for W compared with data from experiments and from the assessment by Gurvich et al. [4].

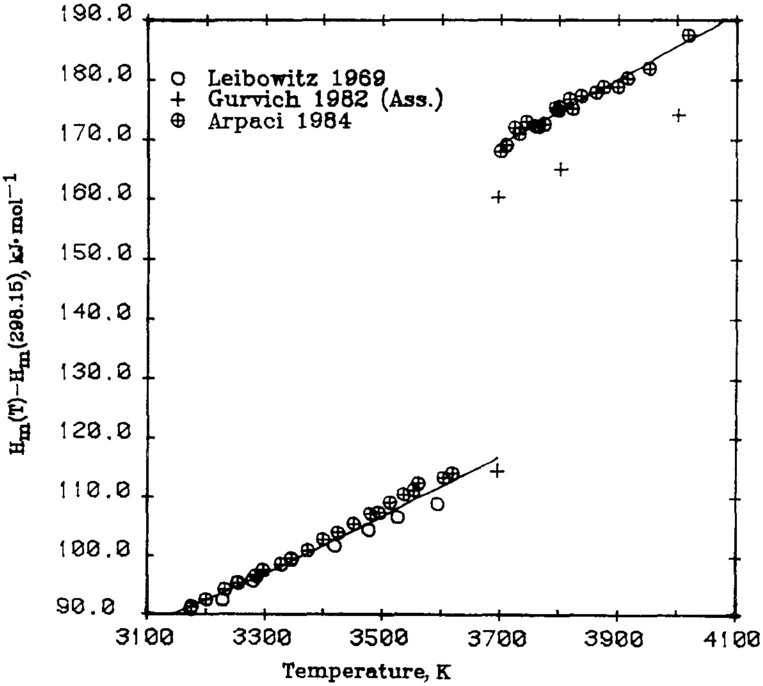

Fig. 2. Calculated enthalpy for W compared with recent data.

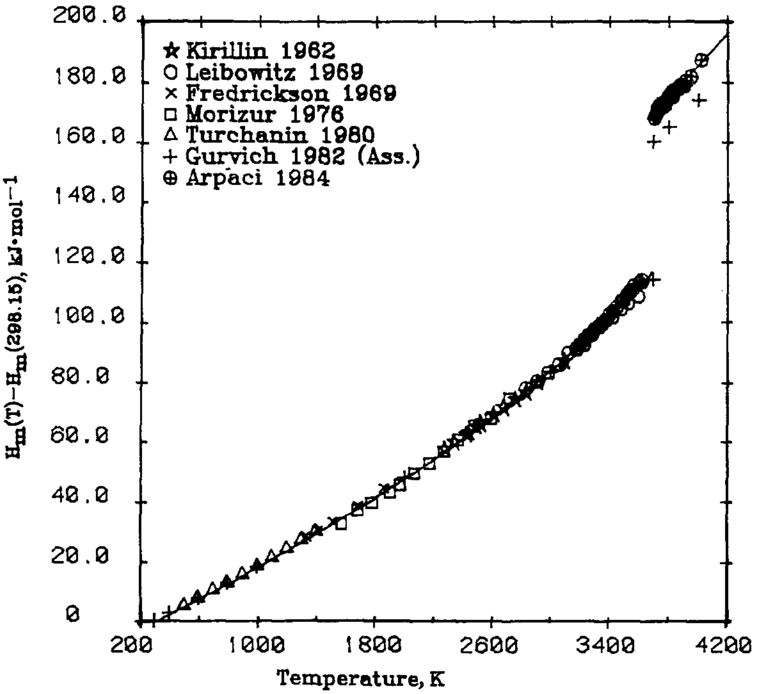

Fig. 3. Calculated enthalpy for W compared with available data.

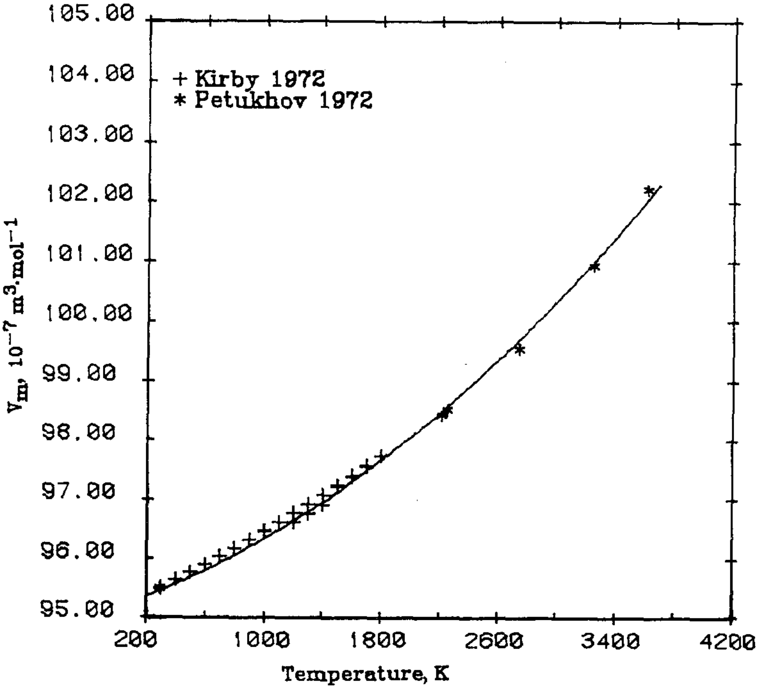

Fig. 4. Calculated molar volume of bcc W versus temperature at 1 atm compared with experimental data.

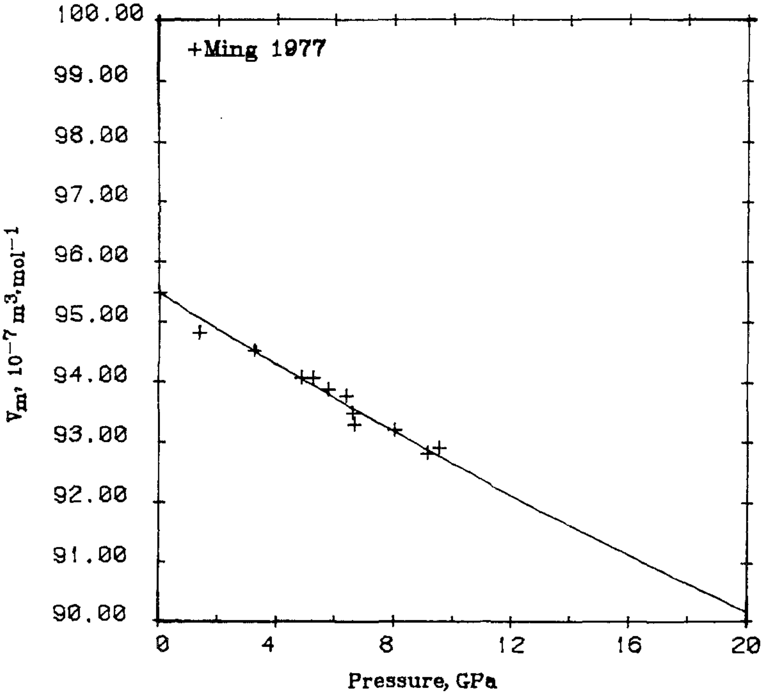

Fig. 5. Calculated molar volume of bcc W versus pressure at 293 K compared with experimental data.

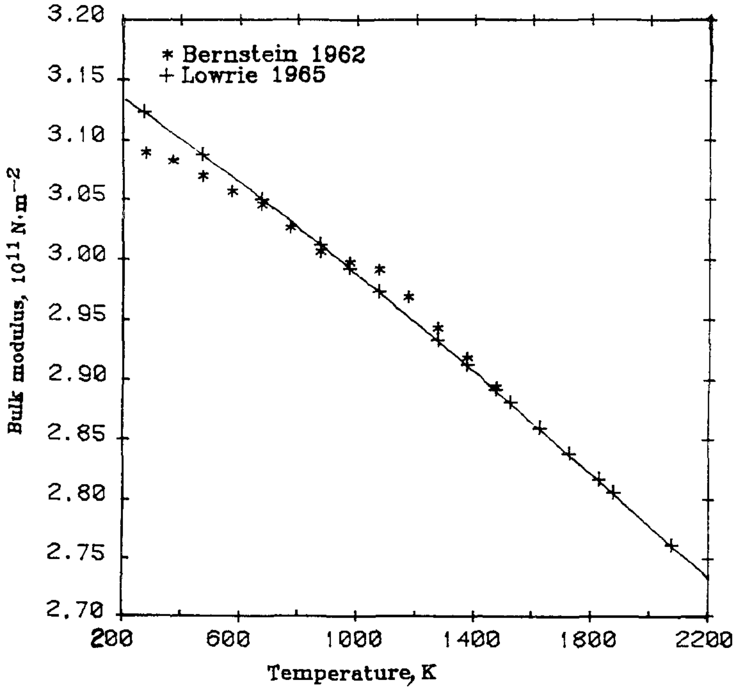

Fig. 6. Calculated bulk modulus versus temperature of bcc W at 1 atm compared with experimental data.

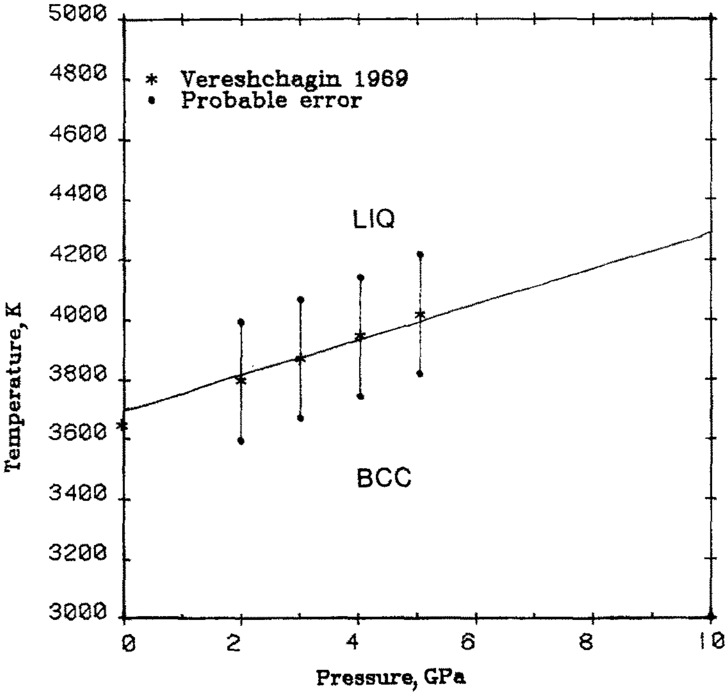

Fig. 7. *P*, *T* phase diagram for tungsten, calculated with the present set of parameters, compared with experimental information.

Figure 3 shows the calculated enthalpies in both solid and liquid W in comparison with calorimetric data [4, 6, 18–23]. The agreement with experimental data above 3100 K is better shown in Fig. 2.

The calculated molar volume of bcc W at atmospheric pressure as a function of temperature is presented in Fig. 4. The agreement is very good.

Figure 5 presents a comparison between the experimental and the calculated effect of pressure on the molar volume of bcc W at 293 K. The agreement is, again, very good.

Figure 6 presents a comparison between the experimental and the calculated temperature dependence of the compressibility at atmospheric pressure expressed by its inverse, the bulk modulus. The agreement with the experimental data of Lowrie and Gonas [14] is excellent.

Figure 7 shows the calculated temperature–pressure phase diagram calculated with the present set of parameters. This phase diagram was obtained by assuming that the thermal expansion and compressibility are the same in liquid as in solid W. The calculated slope of the solid/liquid

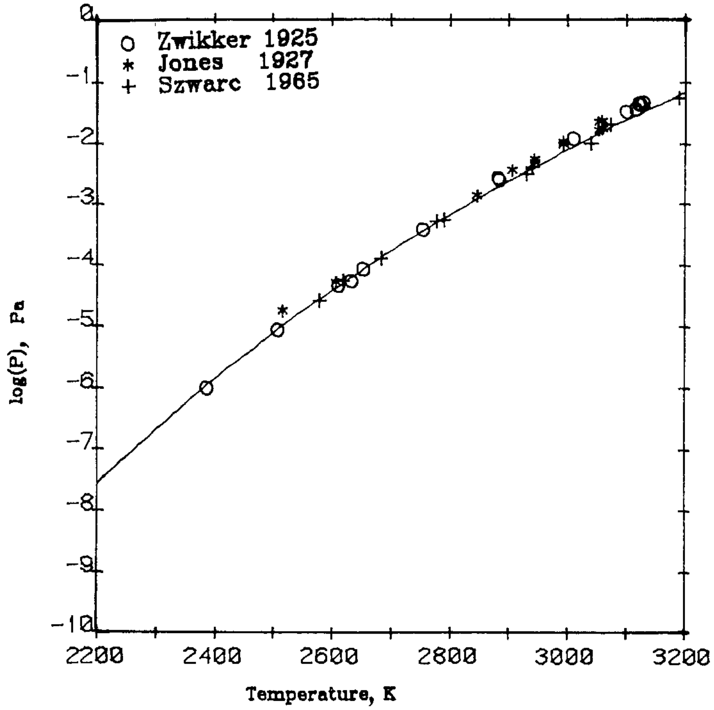

Fig. 8. Calculated vapor pressure of tungsten compared with experimental data.

phase boundary required a volume expansion of 8% at the melting point, which seems to be a realistic value.

Figure 8 shows the calculated vapor pressures of tungsten in comparison with the experimental data quoted in *JANAF* [2]. The calculated heat of sublimation, $\Delta H^0(298.15) = 851.5\ \text{kJ} \cdot \text{mol}^{-1}$, is in good agreement with the values recommended by *JANAF* [2] and Gurvich et al. [4], 851 ± 6 and $850 \pm 5\ \text{kJ} \cdot \text{mol}^{-1}$, respectively.

5. CONCLUSIONS

The results presented in all the diagrams show that a simple equation of state such as the one adopted in this work is capable of describing, within the experimental accuracy, the information available between 298 and 4000 K and between 0 and 10 GPa. However, the empirical nature of the approach and the simplifications introduced in describing the temperature and pressure dependencies of the derivatives of the Gibbs energy restrain the possibilities of making reliable extrapolations outside the range studied experimentally. Such extrapolations can be safe only if models with a sounder physical basis are used. Reasonable extrapolations of the phase boundaries and the thermodynamic properties of the condensed phases may be obtained from the present evaluation up to 6000 K and 40 GPa. Extrapolations to higher temperatures and pressures are dangerous. The simplified description adopted for the molar volume of gaseous W limits the pressure range for reliable calculations including the gas phase to about 20 MPa. The pressure range can be extended by adding additional temrs to Eq. (6). This was not considered worthwhile since a model based on the Helmholtz energy then should be a much better choice.

Full descriptions of all the thermodynamic properties are contained in Tables I–III, and all thermodynamic properties of tungsten can be calculated from these descriptions, preferably by the use of a computer program.

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