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AN ASSESSMENT OF THE THERMODYNAMIC PROPERTIES AND PHASE DIAGRAM OF THE SYSTEM Bi–Cu

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ABSTRACT

The solution thermodynamics and phase equilibria of bismuth–copper alloys at 400–1500 K have been critically analysed. The liquid alloy as well as the copper terminal solution were treated as substitutional alloy phases. Their excess Gibbs energies were fitted by Redlich–Kister polynomials and the optimization of their model parameters was carried out by using the Lukas program. The activities in the liquid alloy deviate positively from Raoult's law, the limiting activity coefficients at the infinite dilution according to the assessment being $\ln f^{\circ}_{[\mathrm{Bi}]\mathrm{Cu}} = 3019/T(\mathrm{K}) - 1.04$ and $\ln f^{\circ}_{[\mathrm{Cu}]\mathrm{Bi}} = 2716/T(\mathrm{K}) - 1.33$. The integral enthalpy of mixing is exothermic and nearly symmetric with a maximum at $x_{\mathrm{Cu}} = 0.52$ with $\Delta H = 6050$ J mol^{-1}. The integral excess entropy of the molten alloy vs. composition was found to be symmetric with the maximum at $\Delta S^{\mathrm{Ex}} = 2.5$ J (mol K)$^{-1}$.

INTRODUCTION

Pure copper crystallizes at 1084.87 °C [1] in an f.c.c. lattice which is stable down to room temperature. Bismuth solidifies at 271.44 °C [1], crystallizing in a rhombohedral form. According to Hansen and Anderko [2] and others [3,4], the binary system of bismuth–copper is of a simple eutectic type with the eutectic point at about 270 °C, close to that of pure bismuth. The solid solubility of bismuth in copper is low, below 0.01 at.% $[\mathrm{Bi}]_{\mathrm{Cu}}$ in maximum [5], and the solubility of copper in solid bismuth is negligible. No stable intermetallic phases have been reported [5].

The phase diagram and the thermodynamic properties of the bismuth–copper binary system have been reassessed by using simultaneously experimental thermodynamic and phase diagram data from original studies for optimizing the adjustable parameters of the solution models. The present evaluation provides supplementary information to the previous critical assessments available in the literature [5–7].

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TABLE 1

The lattice stabilities of pure copper and bismuth used in the calculations [8,9]; reference state SER (J mol^{-1})

Temperature range	A	F	B	I	C	J	D	K	E
Bi (liquid)									
298.15–544.52 K	$3.42816000E+03$	$-8.3815980E-06$	$1.07781519E+02$	$0.000000E+00$	$-2.84096529E+01$	$-5.972600E-19$	$1.23388880E-02$	$0.000000E+00$	$0.0000000E+00$
544.52–800.00 K	$4.15443010E+04$	$1.3499885E-05$	$-4.14461988E+02$	$0.000000E+00$	$5.18556592E+01$	$0.000000E+00$	$-7.53111630E-02$	$0.000000E+00$	$-3.6161680E+06$
800.00–1200.00 K	$2.90615000E+02$	$-1.0460000E-06$	$1.61737334E+02$	$0.000000E+00$	$-3.59824000E+01$	$0.000000E+00$	$7.42660000E-03$	$0.000000E+00$	$0.0000000E+00$
>1200.00 K	$3.75496700E+03$	$0.0000000E+00$	$1.03959803E+02$	$0.000000E+00$	$-2.71960000E+01$	$0.000000E+00$	$0.00000000E+00$	$0.000000E+00$	$0.0000000E+00$
Bi (rho)									
298.15–544.52 K	$-7.81777600E+03$	$-8.3815980E-06$	$1.28418885E+02$	$0.000000E+00$	$-2.84096529E+01$	$0.000000E+00$	$1.23388880E-02$	$0.000000E+00$	$0.0000000E+00$

544.52–800.00 K					
	3.02079400$E+04$	$-3.93650255E+02$	5.18556592$E+01$	$-7.53111630E-02$	$-3.6161680E+06$
	1.3499885$E-05$	0.000000$E+00$	0.000000$E+00$	1.664910$E+25$	
800.00–1200.00 K					
	$-1.10457460E+04$	1.82549067$E+02$	$-3.59824000E+01$	7.42660000$E-03$	0.0000000$E+00$
	$-1.0460000E-06$	0.000000$E+00$	0.000000$E+00$	1.664910$E+25$	
>1200.00 K					
	$-7.58139400E+03$	1.24771535$E+02$	$-2.71960000E+01$	0.00000000$E+00$	0.0000000$E+00$
	0.0000000$E+00$	0.000000$E+00$	0.000000$E+00$	1.664910$E+25$	
Cu (liquid)					
298.15–1358.02 K					
	5.19438200$E+03$	1.20975160$E+02$	$-2.41123920E+01$	$-2.65684000E-03$	5.2478000$E+04$
	1.2922300$E-07$	0.000000$E+00$	$-5.839320E-21$	0.000000$E+00$	
>1358.02 K					
	$-4.69300000E+01$	1.73883734$E+02$	$-3.13800000E+01$	0.00000000$E+00$	0.0000000$E+00$
	0.0000000$E+00$	0.000000$E+00$	0.000000$E+00$	0.000000$E+00$	
Cu (f.c.c.)					
298.15–1358.02 K					
	$-7.77045800E+03$	1.30485403$E+02$	$-2.41123920E+01$	$-2.65684000E-03$	5.2478000$E+04$
	1.2922300$E-07$	0.000000$E+00$	0.000000$E+00$	0.000000$E+00$	
>1358.02 K					
	$-1.35423300E+04$	1.83804197$E+02$	$-3.13800000E+01$	0.00000000$E+00$	0.0000000$E+00$
	0.0000000$E+00$	0.000000$E+00$	0.000000$E+00$	3.646430$E+29$	

$^\circ G_{\mathrm{Bi}}(\mathrm{rom}) - {}^\circ G_{\mathrm{Bi}}(\mathrm{f.c.c.}) = -9900 + 12.50T(\mathrm{K})$

THERMODYNAMIC DESCRIPTION OF THE SYSTEM

Stabilities of the pure components

In the calculations, the analytical form for the temperature dependencies of the Gibbs energies of the pure components according to eqn. (1) was used:

$$^{\circ}G = A + BT + CT \ln T + DT^2 + E/T + FT^3 + IT^4 + JT^7 + KT^{-9} \tag{1}$$

where A, $B, \ldots, K$ are coefficients to be determined from experimental data in the assessment of the elements.

The thermochemical properties of pure bismuth (rho) and pure copper (f.c.c.) at 1 bar were taken from the recent SGTE data file [8]. The coefficients A to K in eqn. (1) for the lattice stabilities of pure bismuth and copper used in the calculations are collected in Table 1. The lattice stability for metastable f.c.c. bismuth was obtained from the same source and it is identical with the recent extensive assessment made by Saunders and Miodownik [9].

Solution models

Pure mathematical solution models of substitutional solutions were adopted to assess the excess Gibbs functions of the molten alloy and the f.c.c. copper terminal solution. Redlich–Kister polynomial expansions for the excess Gibbs energies [10] were used to fit the experimental data.

The general form of the Redlich–Kister polynomials used for the integral excess Gibbs energy of a binary solution phase can be written as:

$$\begin{aligned} {}^{E}G &= x_1 x_2 \left[A_0 + A_1 z + A_2 z^2 + \ldots \right] \\ &= x_1 x_2 \sum A_i (z)^i \end{aligned} \tag{2}$$

where A_i are adjustable temperature- and pressure-dependent parameters and $z = x_1 - x_2 = x_{\mathrm{Bi}} - x_{\mathrm{Cu}}$.

Owing to the scarcity of experimental information, regular solution behaviour was adopted for the f.c.c. copper terminal solution. The solubility of copper in solid bismuth was neglected [5] and it was treated as a stoichiometric phase. The molten copper–bismuth alloy was described by using first-order Redlich–Kister polynomials with temperature-dependent coefficients of the type

$$A_i = A_{i0} + A_{i1} T \tag{3}$$

where A_{i0} and A_{i1} are constants to be estimated from the experimental data and T is the temperature (in K).

EXPERIMENTAL INFORMATION

The phase diagram

The liquidus has been studied independently by several authors, and the experimental information on the phase diagram has been summarized previously by Hansen and Anderko [2] and others [5–7]. The insufficiently documented data of Hiorns [36] were, however, neglected in the calculations, as were the results of Kleppa [34] who reported the solubility data in graphical form only.

Additional equilibrium data were found to be available from the EMF studies made by Lomov and Krestovnikov [11], who reported the original EMF vs. T points for the cell $Cu/Cu^+/[Bi, Cu]$ in tabular form. Because of the low solubility of bismuth in solid copper, linear extrapolation of the

TABLE 2

Experimental data on the bismuth–copper system and the errors used in the optimization

Phase diagram data	Thermodynamic measurements							Ref.
	μ_{Bi}	μ_{Cu}	$\pm d\mu$ (% *)	$\Delta_{mix}H$	$\pm d(\Delta H)$ (J *)	dT ($\pm$K)	dx ($\pm$at.%)	
		+	10			3	1.5	19
	+		20			3	1.5	19
				+	300	3	1.5	19
	+		5			3	1.0	20
	+		200–300 J			3	1.0	21
				+	500	5	2.0	22
				+	40%	3	1.0	23
				+	300	5	1.5	24
+						5	1.5	24
		+	10			2	1.0	25
+						2	0.5	25
+						8	1.5	26
+						1.5	0.5	27
+						3	1.5	28
+						5	1.5	29
+						3	1.5	30
	+		10			3	1.0	31
		+	10			3	1.0	11
+						5	0.5	11
	+		5			2	0.5	12
	+		10			3	0.5	15
	+		15			2	0.25	13
+						3	1.0	33
	+		20			2	0.5	14
+						8	1.5	37

* If not specified otherwise.

results to zero EMF at a constant composition yields the liquidus temperatures.

A large scatter has been reported in the experimental information on the solid solubility of bismuth in copper [5]. The solid solubility of copper in bismuth is below 0.01 at.% $[Cu]_{Bi}$ and, therefore, was neglected in all calculations, bismuth being treated as a pure stoichiometric phase.

The experimental phase-diagram points available in the primary sources are collected in Table 2 which also shows the uncertainties of each study adopted in the final parameter optimization.

Thermodynamic properties

The thermodynamic studies reported up to 1982 have been collected by Chakrabarti and Laughlin [5]. No thermodynamic data appear to be available on the terminal copper solid solution. Since the 1982 survey [5], only a few experimental papers have been published on the copper–bismuth binary system. Sibanda and Baker [12], Arac and Geiger [13] and Roine and Jalkanen [14] used transportation techniques to determine the activity coefficient of bismuth at 1100–1250 °C in dilute copper-rich alloys. Itoh and Azakami [15] used the Knudsen cell technique combined with a microbalance at 1100 °C. Jiang et al. [16] have recently reported additional data for the activity coefficient of bismuth in molten copper alloys at low bismuth concentrations.

The experimental thermodynamic data and their inaccuracies used in the optimization of the model parameters are collected in Table 2. Owing to the limited enthalpy of mixing data on the molten alloys also the mixing enthalpies derived from the EMF results of Nikolskaya et al. [19] were included in the experimental file.

OPTIMIZATION OF THE MODEL PARAMETERS

The program BINGSS [17,18] (the double-precision version updated on 27 July 1989) was used for the least-squares optimization of the coefficients of the excess Gibbs-energy functions of the alloy phases. The phase diagrams were calculated by using the program BINFKT (the version updated on 16 August 1989). The calculations were carried out on an IBM 3090-150 computer at the Computer Centre of Helsinki University of Technology.

The assessed phase diagram

The assessed phase diagram for the temperature interval 400–1500 K together with the experimental observations from the literature are shown in Fig. 1. The equilibrium data on the phase diagram [11,25,27,33] (Table 2)

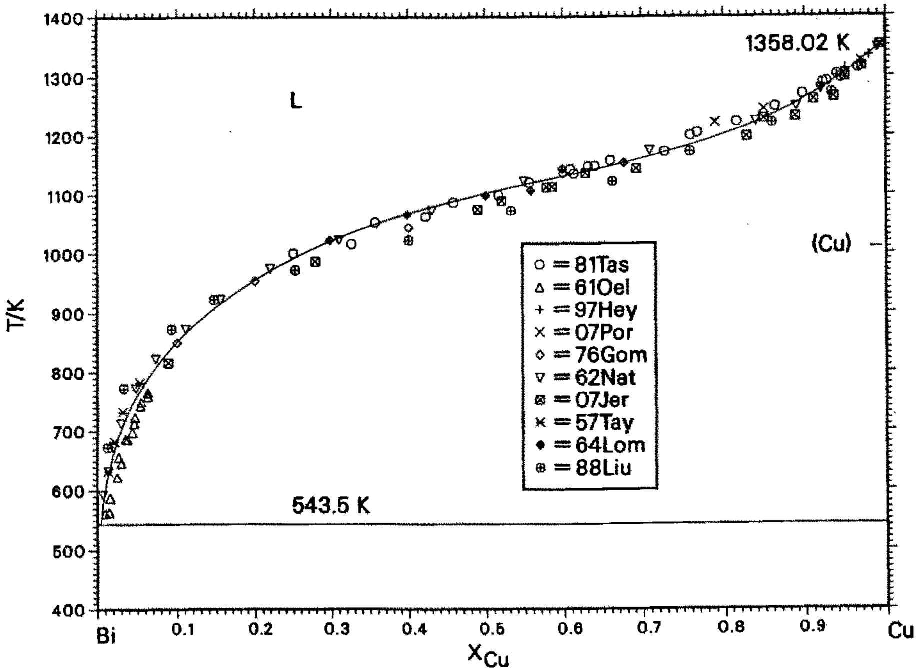

Fig. 1. The calculated phase diagram for bismuth–copper at 400–1400 K: 97Hey [30], 07Jer [26], 07Por [29], 57Tay [33], 61Oel [24], 62Nat [27], 64Lom [11], 76Gom [28], 81Tas [25], 88Liu [37], (———) calculated (this work).

were preferred in the optimization to the dynamic studies [24,29,30] by giving them a smaller inaccuracy. The assessed liquidus is in good agreement with the solubilities carefully measured by Nathans and Leider [27], obtained by a static sampling technique over the whole concentration interval. The results are also in good agreement with the EMF data of Lomov and Krestovnikov [11] and Taskinen and Niemelä [25].

The detail of the phase relations at low temperatures, in the vicinity of the eutectic point, is reproduced in Fig. 2. As a result of the least-squares optimization, the eutectic equilibrium was calculated to be formed at 270.3° C (543.5 K), 1.2 K below the melting point of pure bismuth, the location of the eutectic point being ${}^{e}x_{Cu} = 0.0047$. The assessed eutectic point is in fair agreement with the experimental value of ${}^{e}x_{Cu} = 0.0048$ [34]; Nathans and Leider [27] reported ${}^{e}t = 270.6 \pm 0.1°$ C at ${}^{e}x_{Cu} = 0.0046$ and Jeriomin [26] obtained ${}^{e}t = 270 \pm 2°$ C at ${}^{e}x_{Cu} = 0.005$. The recent critical evaluations of Chakrabarti and Laughlin [5] and Niemelä et al. [7] gave ${}^{e}t = 270.5°$ C at ${}^{e}x_{Cu} = 0.005$ and ${}^{e}t = 270.6°$ C at ${}^{e}x_{Cu} = 0.006$, respectively; these values are in good agreement with the result of the present work. Figure 2 indicates that the liquidus temperatures measured by Oelsen et al. [24] in bismuth-rich alloys show systematic undercooling if compared with the results of Nathans and Leider [27] and Taylor [33] or with the calculated line.

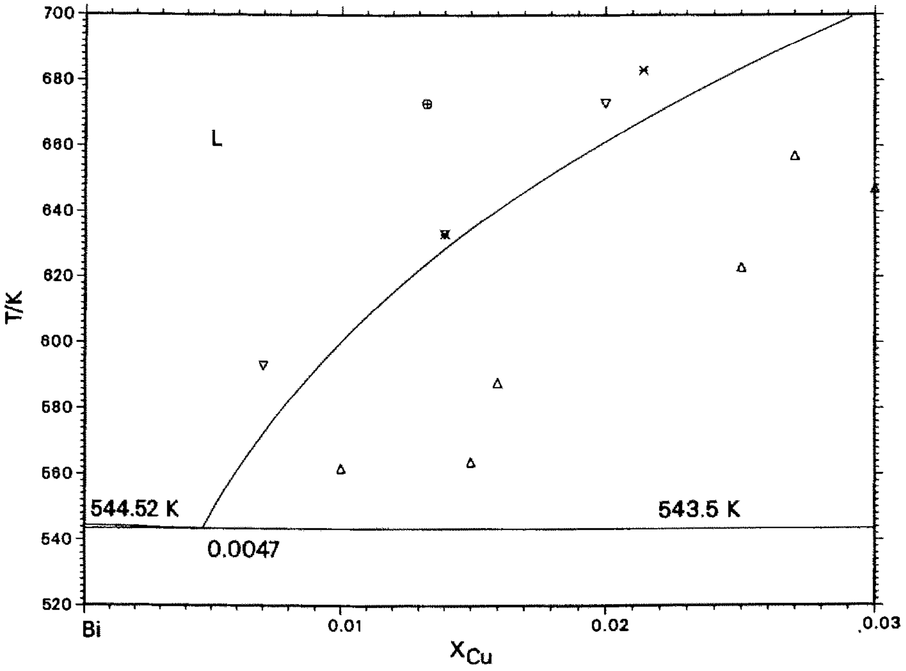

Fig. 2. Detail of the phase diagram in the vicinity of the eutectic point: the calculated eutectic point is at ${}^{e}t = 543.5$ K (270.2° C) and ${}^{e}x_{\mathrm{Cu}} = 0.0047$. Symbols as in Fig. 1.

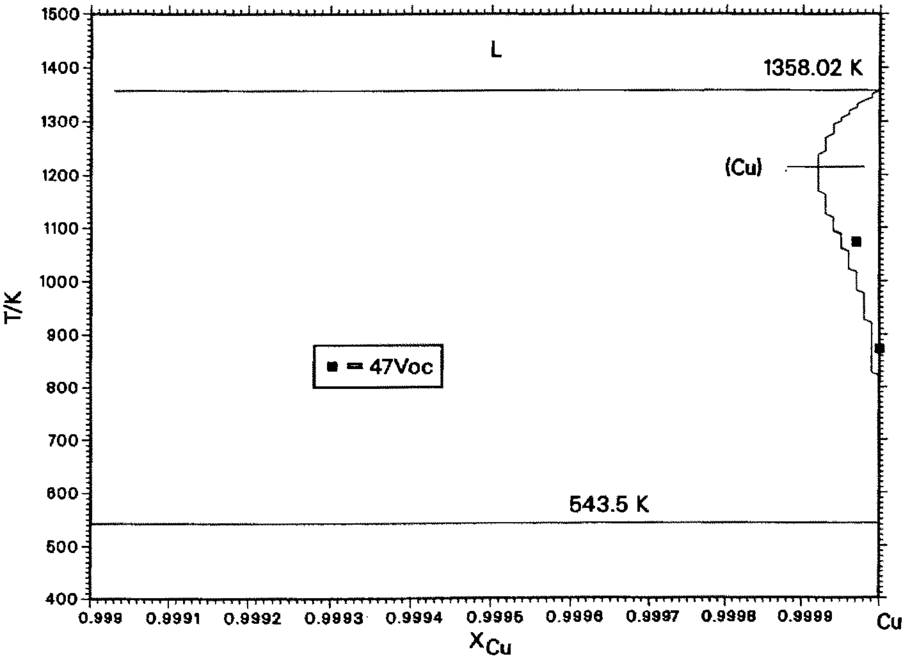

Fig. 3. The terminal copper region at 400–1500 K: 47Voc [32]; (———) calculated (this work).

The calculated retrograde shape of the terminal copper solid solution and the experimental points used in the calculations are shown in Fig. 3. Owing to the significant scatter in the experimental data [5] the solubilities measured by Voce and Hallowes [32] and recommended by Chakrabarti and Laughlin [5] were selected for the optimization. As one can see, the maximum solubility of bismuth in solid copper is very low and at the eutectic temperature the solubility is less than 0.001 at.% $[\mathrm{Cu}]_{\mathrm{Bi}}$.

Solution thermodynamics

The optimized coefficients of the thermodynamic excess functions of the alloy phases obtained in this work are listed in Table 3. Figure 4 shows the estimated integral enthalpy of mixing of molten bismuth–copper alloys together with the experimental points and the values selected by Hultgren et al. [6]. The standard states were Cu(l) and Bi(l). The integral enthalpy of mixing is nearly symmetric with the maximum lying at about $x_{\mathrm{Cu}} = 0.52$ with $\Delta H = 6050$ J mol^{-1}. In this assessment, the temperature dependence of the enthalpy of mixing for the molten alloy was assumed to be zero by adopting $\Delta c_p = 0$. The values of Kawakami [22] are highly scattered and the accuracy achieved for them in the final optimization was as low as ± 500 J mol^{-1}. The assessed enthalpy of mixing is in good agreement with the

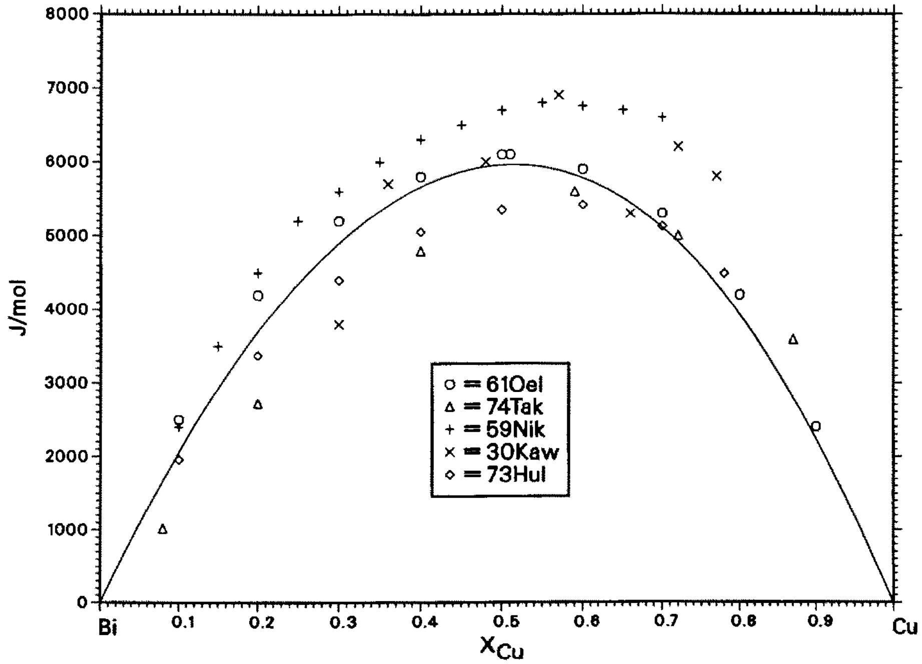

Fig. 4. Integral enthalpy of mixing of the molten bismuth–copper alloys: 30Kaw [22]; 59Nik [19]; 61Oel [24]; 71Hul [6]; 74Tak [23]; (———) this work; standard states Bi(l) and Cu(l).

TABLE 3

The estimated Redlich–Kister parameters A_{ij} for Bi–Cu alloys (J mol^{-1})

$i\backslash j$	0	1
Liquid		
0	23844.75	−9.84341
1	−1260.32	−1.19289
f.c.c.		
0	77868.73	0

observations of Oelsen et al. [24] as well as with the values recommended by Niemelä et al. [7]. The mixing enthalpies of Nikolskaya et al. [19] calculated from their EMF vs. T data are too exothermic by about 1 kJ mol^{-1}.

The assessed entropy and excess entropy of mixing of the molten alloy is shown in Fig. 5. The graph also compares the corresponding values at 1200 K suggested by Hultgren et al. [6] with the optimized, temperature-independent curve. It can be seen that agreement with Hultgren's data is fair and the entropy vs. composition curves are essentially symmetric with the maximum at $\Delta S^{\mathrm{Ex}} = 2.5$ J (mol K)$^{-1}$.

The calculated activities of bismuth and copper at 1200 K and at 1400 K in the molten alloy according to the present assessment, with liquid bismuth

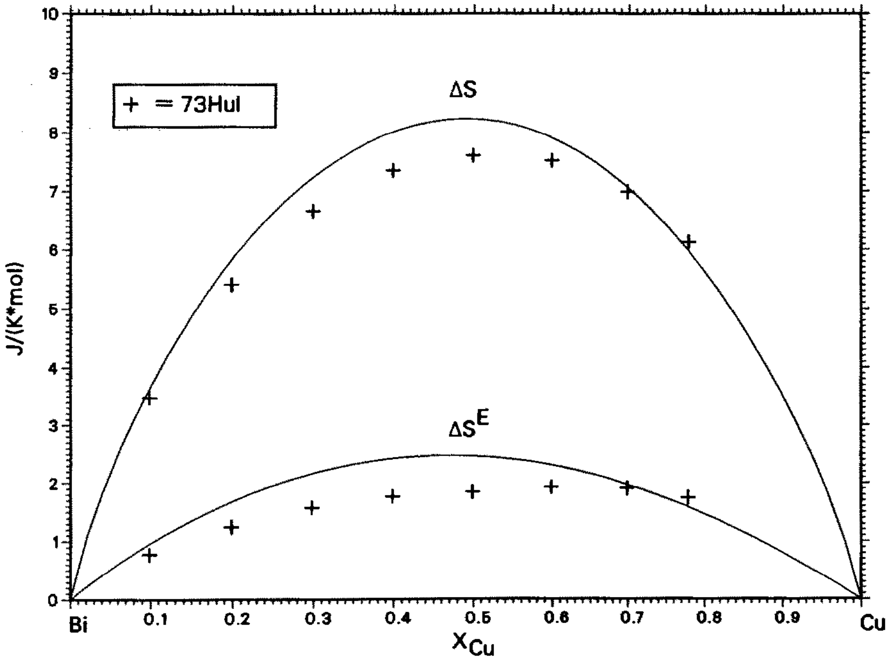

Fig. 5. The integral entropy of mixing and the integral excess entropy of mixing: 71Hul [6]; (———) this work; standard states Bi(l) and Cu(l).

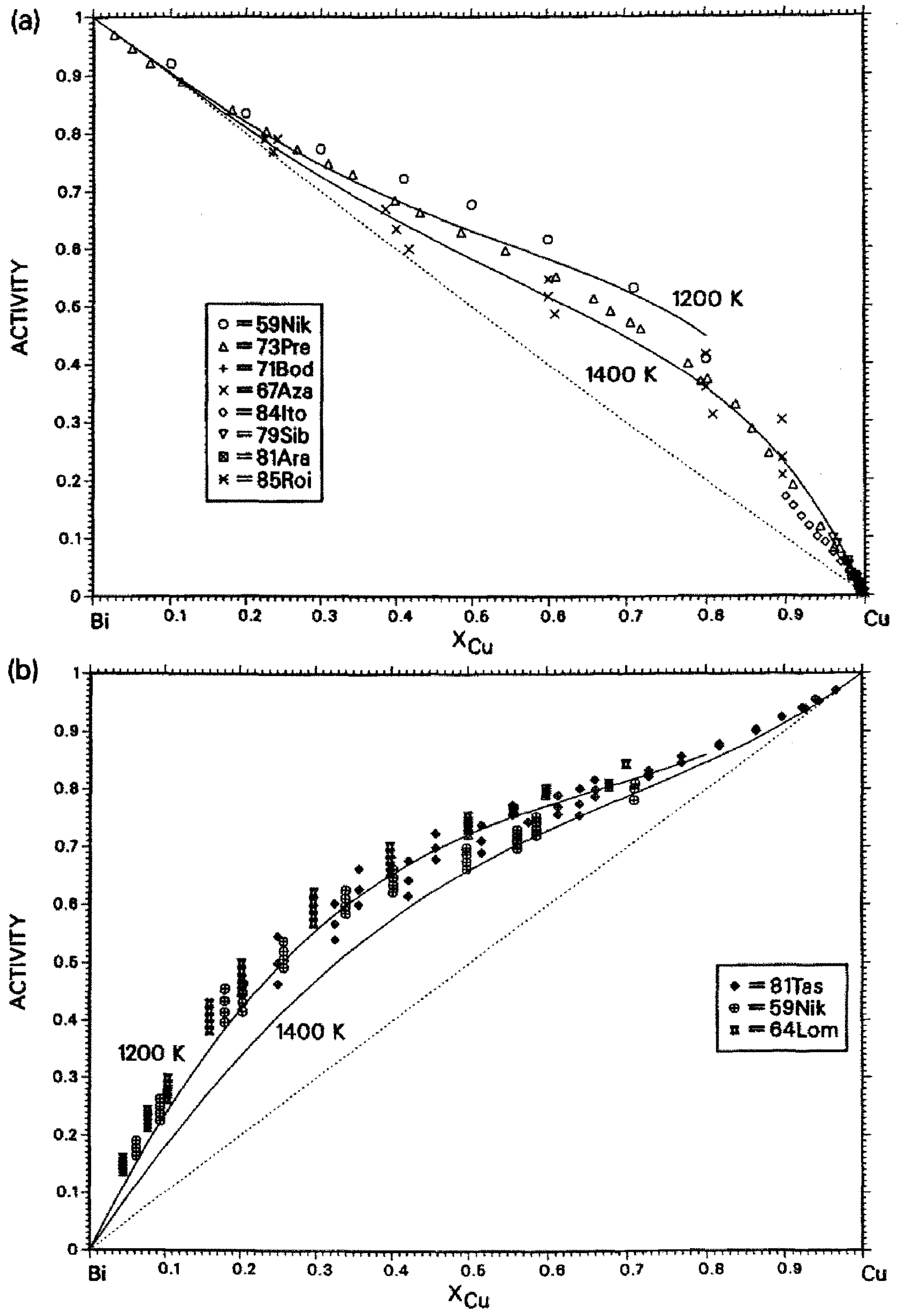

Fig. 6. Activity of bismuth (a) and copper (b) in the liquid alloy: 59Nik [19]; 64Lom [11]; 67Aza [31]; 71Bod [20]; 73Pre [21]; 79Sib [12]; 81Ara [13]; 81Tas [25]; 84Ito [15]; 85Roi [14]; (———) this work at 1200 K and 1400 K; (· · · · · ·) Raoultian solution; standard states Bi(l) and Cu(l).

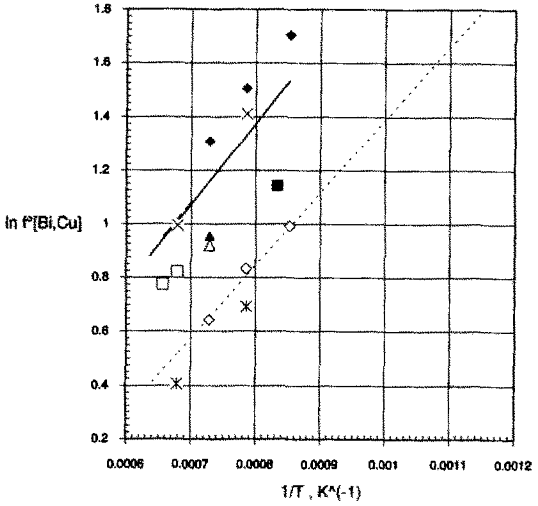

Fig. 7. The activity coefficients of bismuth and copper at infinite dilution in liquid bismuth copper alloys as a function of the inverse absolute temperature: ■, 59Nik [19] Cu; × and ⋊, 67Aza [31] Bi and Cu; ▲, 79Sib [12] Bi; ◆ and ◇, 81Tas [25] Bi and Cu; △, 84Ito [15] Bi; — — — —, 85Kel [35] Bi; ———, ln $f^{\circ}_{[\mathrm{Bi}]\mathrm{Cu}}$ and ······, ln $f^{\circ}_{[\mathrm{Cu}]\mathrm{Bi}}$ assessed in this work; standard states Bi(l) and Cu(l).

and copper as the standard states, are shown in Fig. 6 as a function of the composition of the melt. The experimental points are also reproduced in the graph. The Raoultian solution is shown in the graph by the dotted line. In general, all the experimental activities for copper and bismuth available in the literature are in good agreement, with the exception of those reported by Itoh and Azakami [15]. These authors reported systematically too low bismuth activity values in copper-rich alloys at 1373 K by using a Knudsen effusion technique combined with an electrobalance.

The limiting activity coefficients of bismuth in molten copper and copper in molten bismuth at infinite dilution are frequently needed in qualitative calculations for engineering purposes. The limiting activity coefficients at infinite dilution according to the present assessment, obtained from the optimized set of coefficients given in Table 3, are

$$\left.\begin{array}{l} RT \ln f^{\circ}_{[\mathrm{Bi}]\mathrm{Cu}} = 25\,105.1 - 8.651T \\ \text{and} \\ RT \ln f^{\circ}_{[\mathrm{Cu}]\mathrm{Bi}} = 22\,584.4 - 11.036T \end{array}\right\} \qquad (4)$$

where T is in K. In eqn. (4) the standard states of the dissolved elements are Bi(l) and Cu(l). The available experimental values are shown graphically in Fig. 7. The ln f° vs. $1/T$ plot also compares the available experimental primary data as well as the estimated activity coefficient of bismuth from Kellogg et al. [35] with the calculated lines.

SUMMARY

The Lukas least-squares optimization program has been applied to calculating the Redlich–Kister parameters of the excess Gibbs energies of the alloy phases in the bismuth–copper binary system over the temperature interval 400–1500 K. The liquid alloy and the narrow fcc copper terminal solution were treated as solution phases.

An internally consistent set of thermodynamic excess functions using Redlich–Kister polynomials was obtained from the optimization procedure by simultaneously using the experimental phase diagram and the thermodynamic data of the system. The calculated phase diagram and the necessary thermodynamic model parameters are reported.

The integral excess Gibbs energy (in J mol^{-1}) of mixing the molten alloy is expressed as

$$\Delta G^{\mathrm{Ex}} = x_{\mathrm{Bi}} x_{\mathrm{Cu}} [23\,845 - 9.843T - (1\,260 + 1.193T)(x_{\mathrm{Bi}} - x_{\mathrm{Cu}})]$$

The activities of the component metals deviate positively from a Raoultian solution, the limiting activity coefficients at infinite dilution in the molten alloy being $\ln f^{\circ}_{[\mathrm{Bi}]\mathrm{Cu}} = 3019/T(\mathrm{K}) - 1.04$ and $\ln f^{\circ}_{[\mathrm{Cu}]\mathrm{Bi}} = 2716/T(\mathrm{K}) - 1.33$.

The solid solubilities of bismuth in copper and copper in bismuth are very low, and a maximum solubility of bismuth at about 1200 K with ${}^{\star}x_{[\mathrm{Bi}]} = 0.0001$ was estimated.

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