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A THERMODYNAMIC EVALUATION OF THE Au–Sn SYSTEM

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ABSTRACT

Available experimental information concerning thermodynamic properties as well as phase equilibria data has been compiled and used for the assessment of new self-consistent parameters of thermodynamic models for all phases in the Au–Sn system. These coefficients have been optimized by using a program developed by Lukas et al. Calphad, 1 (1977) 225.

Phase diagram and characteristic thermodynamic functions have been calculated and compared with the experimental values using the THERMODATA software. A very satisfactory agreement has been obtained by using twenty-eight optimized coefficients for about five hundred experimental points, with a simple substitutional model for solution phases and a linear approximation for the temperature dependency of the Gibbs energy of the stoicheiometric compounds.

INTRODUCTION

During recent years, thermodynamic calculations of the state of equilibrium in multicomponent systems have taken on increasing importance, in order to solve industrial problems encountered most of the time in the field of materials science [2,3]. The main advantage of this approach is to limit the experimental investigation field by predicting, as a first step, some expected equilibrium state of the system for any given temperature, composition or partial pressure provided by the user and, in a second step, by improving the theoretical approach with some well-selected experiments and eventually by including multicomponent interaction terms.

People involved with this kind of approach were soon coming up against the lack of self-consistent data concerning the basic systems, especially unitary or binary systems.

Indeed, although there are many publications concerned with the basic systems, experimental data are often from different sources, and not always self-consistent. Moreover, some fields of a system may have been extensively studied by many authors whereas some other fields are still unexplored.

In order to develop a solution to these two main problems, i.e. the lack of self-consistent data for basic systems and duplication of effort, international

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cooperation has been organized inside European groups (like S.G.T.E. = Scientific Group Thermodata Europ) in order to produce data for selected systems and to supply specialized data banks.

Within this framework, critical assessment of the gold–tin system is developed, and this work is a part of a much more important project concerning twenty-five binary systems.

SHORT PRESENTATION OF THE DIFFERENT PHASES

The assessed phase diagram, reported by Okamoto and Massalski [4], shows the existence of numerous solution phases and stoicheiometric compounds which are listed below, with the symbols currently used.

Solution phases

(1) liquid phase (L)
(2) terminal rich gold solid solution, with a face-centered cubic (A1) prototype structure: (fcc)
(3) terminal rich tin solid solution, with a body-cubic tetragonal (A5) prototype structure: (bct)
(4) terminal rich tin solid solution, with fcc (A4) structure isotopye with diamond: (dia)
(5) intermediate solid solution, with a Mg-type hcp (A3) structure: (hcp or ζ)

Intermetallic compounds

(6)	$Au_{10}Sn(\beta)$	Ni_3Ti prototype
(7)	$Au_5Sn(\zeta')$	hcp symmetry
(8)	$AuSn(\delta)$	AsNi prototype (homogeneity range between 50 and 50.5 at% Sn)
(9)	$AuSn_2(\varepsilon)$	orthorhombic structure
(10)	$AuSn_4(\eta)$	$PtSn_4$ prototype

EXPERIMENTAL INFORMATION

Phase diagram

Phase equilibria of the Au–Sn system have been experimentally determined using conventional techniques by Vogel [5] (liquidus, invariant; thermal, microscopic), Haycock and Neville [6] (liquidus; thermal), Stenbeck and Westgren [7] (solidus/solvus; lattice parameter), Jenckel and Roth [8] (solidus/solvus; resistivity, etc.), Owen and Roberts [9] (solidus/solvus; lattice parameter), Schubert et al. [10] (solidus/solvus; X-ray), Massalsky and King [11] (solidus/solvus; X-ray), Misra et al. [12] (liquidus; thermal), Prince [13] (liquidus; thermal), Davies and Leach [14] (liquidus; thermal), Petot et al. [15] (liquidus; e.m.f.), Osada et al. [16] (solidus/solvus, invariant; specific heat), Hayer et al. [17] (liquidus; enthalpy of mixing), Itagaki [18] (liquidus; thermal), Wallbrecht et al. [19] (liquidus, invariant; specific heat), Vnuk et al. [20] (solvus; hardness), and Legendre et al. [21] (solidus/solvus, invariant; thermal).

Experimental values from the different authors have been compiled using the up-dating of Okamoto and Massalski [4] as well as the original articles, and entered in a file which can be used by the optimization procedure.

Moreover, the recent study of Legendre et al. [21] has allowed us to take into account more accurately the different phase equilibria between the different solution phases (L, ζ, fcc) and compounds (β, ζ') in the gold rich region (Sn < 25 at%).

Thermodynamic properties

A bibliographic search on thermodynamic properties of the Au–Sn system was made by means of the "Thermdoc bibliographic data base" integrated in the THERMODATA system [22].

Intermetallic compounds and solid solutions

Values of enthalpies of formation ΔH (J per gram atom) have been measured by calorimetry by Misra et al. [12] ($T = 273$ K, ζ, δ, ε, η); Kleppa [23] ($T = 723$ K, fcc, ζ; $T = 525$ K, ε, η); Jena and Bever [24] ($T = 273$, 195, 78 K, δ, ζ); Masse et al. [25] ($T = 696$ K, δ). Values of enthalpies and entropies of formation in the temperature range 298.15–691 K have been estimated by Hultgren et al. [26]. The heat of fusion of AuSn(δ) has been also measured by Misra et al. [12] and Kubaschewski [27].

The specific heats of AuSn(δ) determined by Misra et al. in the temperature range 613–673 K are appreciably higher than those of Bottema and Jaeger [28] or Hultgren et al. [26] but in reasonable agreement with the equation given by Kelley [29] which is based mainly on the results of Kubaschewski [27].

The specific heat of $\mathrm{AuSn_4}(\eta)$ has been determined using DSC by Wallbrecht et al. [19], and the following equation is proposed

$$Cp(\eta) = 21.0 + (16.6 \times 10^{-3})T + 47324\ T^{-2}$$

for $298.15 < T < 480$ K.

The partial Gibbs energy of Sn in the fcc solid solution has been measured using the gas–solid equilibration technique of Alcock and Jacob [30] ($T = 1000$ K, $x_{\mathrm{Sn}} = 0.01$–0.04) and by the e.m.f. method of Predel and Schallner [31] ($T = 1000$ K, $x_{\mathrm{Sn}} = 0.01$–0.04).

Liquid phase

Values of the partial Gibbs energy of Sn in the liquid phase have been measured using the e.m.f. method of Elliott and Lemons [32] ($T = 541$ K, $x_{\mathrm{Sn}} = 0.906$–0.999), Kleppa [33] ($T = 873$ K, $x_{\mathrm{Sn}} = 0.222$–0.903), Petot et al. [15] ($T = 623$–1323 K, $x_{\mathrm{Sn}} = 0.10$–0.95), Kameda et al. [34] ($T = 944$–1256 K, $x_{\mathrm{Sn}} = 0.17$–0.90), and by mass spectrometry by Howard and Hager [35] ($T = 1873$ K, $x_{\mathrm{Sn}} = 0$–1).

Values of enthalpy of mixing ΔH have been determined using calorimetry by Jena and Leach [36] ($T = 548, 573, 623, 673, 723$ K, $x_{\mathrm{Sn}} = 0$–1), Kleppa [37] ($T = 515, 543, 623, 723$ K, $x_{\mathrm{Sn}} = 0.8$–1), Itagaki et al. [38] ($T = 1373$ K, $x_{\mathrm{Sn}} = 0$–1), Pool et al. [39] ($T = 1380$ K, $x_{\mathrm{Sn}} = 0.142$–1), Hayer et al. [17] ($T = 630$–1695 K, $x_{\mathrm{Sn}} = 0$–1), Rakotomavo [40] ($T = 1373$ K, $x_{\mathrm{Sn}} = 0$–1), Masse [41] ($T = 696$ K, $x_{\mathrm{Sn}} = 0$–1), using an e.m.f. method by Petot et al. [15] ($T = 623$–1323 K, $x_{\mathrm{Sn}} = 0.10$–0.95), Kleppa [33] ($T = 823$ K, $x_{\mathrm{Sn}} = 0$–1), Kameda et al. [34] ($T = 1223$ K, $x_{\mathrm{Sn}} = 0$–1), and using mass spectrometry by Howard and Hager [35] ($T = 1873$ K, $x_{\mathrm{Sn}} = 0$–1).

EVALUATION METHOD

The Gibbs energy of different solution phases has been described using a simple substitution model. An expression for the excess Gibbs energy has been developed by a Redlich-Kister polynomial equation of the form

$$\Delta^{\mathrm{E}}G = x_{\mathrm{Au}} x_{\mathrm{Sn}} \sum_{\nu=0}^{n} (x_{\mathrm{Au}} - x_{\mathrm{Sn}}) L^{(\nu)}_{\mathrm{Au,Sn}}(T) \tag{1}$$

where $L^{(\nu)}_{\mathrm{Au,Sn}}(T) = a^{(\nu)}_{\mathrm{Au,Sn}} + b^{(\nu)}_{\mathrm{Au,Sn}} T$ and the interaction terms $L^{(\nu)}_{\mathrm{Au,Sn}}(T)$ are linear functions of the temperature, i.e. $a^{(\nu)}_{\mathrm{Au,Sn}}$ and $b^{(\nu)}_{\mathrm{Au,Sn}}$ correspond to the temperature-independent values of the enthalpy and the excess entropy of mixing. This assumption has been made because the variation of the experimental enthalpy of mixing with temperature is too complex to be taken into account. Moreover, a temperature dependence of ΔH and $\Delta^{\mathrm{E}}S$ should certainly need more complicated thermodynamic models to take into account the existence of short range order or associate formation in liquid Au–Sn alloys, for which some doubts still remain. Finally, these complex phenomena in liquid alloys have been extensively discussed by Jena and Leach [36], Chatillon-Colinet et al. [42], Janitsch et al. [43] and Hayer et al. [17] without reaching a definitive conclusion.

For all the stoicheiometric compounds, the Gibbs energy of formation has been represented by a linear expression

$$\Delta G = a + bT \tag{2}$$

No dependence on temperature has been considered for the enthalpy and the entropy of formation of stoicheiometric compounds, because not enough accurate information exists for the values of the heat capacity of the compounds, except for $AuSn_4(\eta)$, which has been determined by Wallbrecht et al. [19]. But even for this compound, the calculated values for ΔC_p (using the S.G.T.E. values for pure Au and Sn) were equal to less than -0.3 J per gram atom in the temperature range 298.15–480 K; this is too small to be taken into account. The compound AuSn shows a narrow homogeneity range, and has been considered as stoicheiometric.

The values used for the lattice-stabilities of the pure components come from the S.G.T.E. assessment [44] for the stable solid and liquid phases and from the assessment of Saunders [45] for the metastable phases (required to express the excess properties of mixing of the different solid solution phases), except for the bct structure of gold, which has been estimated arbitrarily as 4184 J.

The following values have been used [44,45]

$$G_{Au}^{\ominus L} - G_{Au}^{\ominus fcc} = 12589.413 - 9.416744T + (8140.249 \times 10^{-25})T^7$$

for $298.15 < T < 1337.58$ K

$$= 12522.901 - 9.364532T + (398805.149 \times 10^{23})T^{-9}$$

for $1337.58 < T < 3200.0$ K

$$G_{Au}^{\ominus hcp} - G_{Au}^{\ominus fcc} = 550 + 0.30T$$

$$G_{Au}^{\ominus bct} - G_{Au}^{\ominus fcc} = 4184 \quad \text{[estimation]}$$

$$G_{Sn}^{\ominus L} - G_{Sn}^{\ominus bct} = 7104.380 - 14.089569T + (14950281.636 \times 10^{-25})T^7$$

for $298.15 < T < 505.12$ K

$$= 6970.585 - 13.811447T + (125.305 \times 10^{23})T^{-9}$$

for $505.12 < T < 3000.0$ K

$$G_{Sn}^{\ominus fcc} - G_{Sn}^{\ominus bct} = 4150 - 5.2T$$

$$G_{Sn}^{\ominus hcp} - G_{Sn}^{\ominus bct} = 3900 - 4.4T$$

The different parameters of the various solution phases $a_{Au,Sn}^{(\nu)}(\phi)$ and $b_{Au,Sn}^{(\nu)}(\phi)$, and stoicheiometric compounds $a(\phi)$ and $b(\phi)$ have been optimized by using a program developed by Lukas et al. [1] which takes into account all the available experimental information and accuracy, after having discarded by a primary criticism some values either for experimental reasons or because they were too indirectly measured (ϕ is the symbol for any phase of the system).

RESULTS AND DISCUSSION

Tables 1 and 2 show the optimized parameters of the excess Gibbs energy for various solution phases referred to the pure elements having the same

TABLE 1

Excess Gibbs energy, $\Delta^E G$, coefficients for the liquid, fcc, hcp(ζ), bct phases of the Au–Sn system.

$$\Delta^E G = x_{Au} x_{Sn} \sum_{\nu=0}^{n} (x_{Au} - x_{Sn}) L^{(\nu)}_{Au,Sn}(T)$$

$$L^{(\nu)}_{Au,Sn}(T) = a^{(\nu)}_{Au,Sn} + b^{(\nu)}_{Au,Sn} T \quad (\nu = 0, 1, 2, 3)$$

Phase	ν	$a^{(\nu)}_{Au,Sn}$ (J)	$b^{(\nu)}_{Au,Sn}$ (J K^{-1})
liquid	0	−47403.1	−3.24717
	1	−15273.5	−3.20514
	2	−10437.3	+9.30294
	3	−11770.0	+13.03362
fcc	0	−8438.7	−20.82759
	1	−12351.5	0
hcp or ζ	0	−19286.9	+17.40660
	1	−50263.4	−41.24114
	2	44703.0	0
bct	0	−7191.9	−22.67792

structure as the corresponding phase, and of the Gibbs energy of formation of the stoechiometric compounds referred to Au(fcc) and Sn(bct).

The phase diagram was calculated using the optimized coefficients and compared with the experimental one in Fig. 1. Table 3 shows experimental and calculated temperature and composition for the invariant equilibria and temperatures of fusion or decomposition of stoicheiometric compounds.

Very good agreement is observed for all the invariant temperatures, except for the reaction $Au_{10}Sn + L \rightleftharpoons$ hcp, which is lower than the experimental one reported in refs. 21. But it must be kept in mind that the phase equilibria between L, fcc, hcp (ζ) and $Au_{10}Sn$ in the gold-rich region are not still completely established and that there is disagreement between the types

TABLE 2

Gibbs energy, ΔG, coefficients for the stoicheiometric compounds β, ζ', δ, ε, and η, referred to as Au(fcc) and Sn(bct).

$\Delta G = a + bT$

Compound	x_{Sn}	a	b
$Au_{10}Sn(\beta)$	0.091	−1546.8	−4.29185
$Au_5Sn(\zeta')$	0.167	−4200.0	−4.05185
$AuSn(\delta)$	0.500	−15144.2	+0.68990
$AuSn_2(\varepsilon)$	0.667	−14217.1	+4.74976
$AuSn_4(\eta)$	0.800	−7439.7	−0.04366

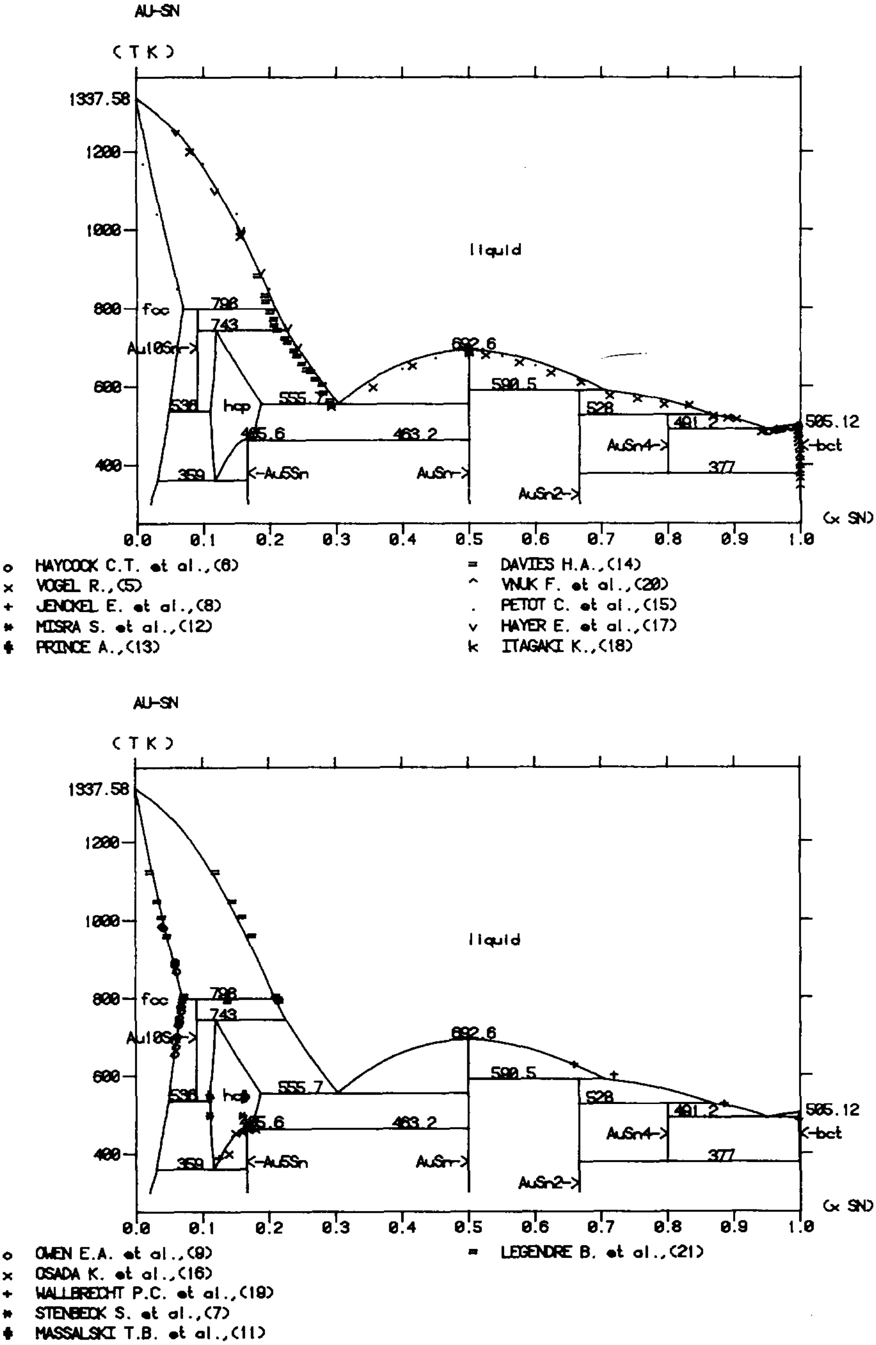

Fig. 1. Comparison between the calculated Au–Sn phase diagram and selected experimental data.

TABLE 3

Three phase equilibria and congruent transformations in the Au–Sn system

Transformation	Calculated		Experimental		Reference
	T (K)	$x = x_{Sn}$	T (K)	$x = x_{Sn}$	
$AuSn_4 \rightleftharpoons AuSn_2 + bct$	377	$x^{bct} = 0.9997$	323	(dashed points)[1]	4
$L \rightleftharpoons AuSn_4 + bct$	491	$x^{bct} = 0.9966$ $x^{L} = 0.949$	490	$x^{bct} = 0.998$ $x^{L} = 0.937$	4, 5
			488		19
$AuSn_2 + L \rightleftharpoons AuSn_4$	527.8	$x^{L} = 0.867$	525	$x^{L} = 0.885$	4, 5
			527, 529		19
$AuSn + L \rightleftharpoons AuSn_2$	590.5	$x^{L} = 0.705$	582	$x^{L} = 0.72$	4, 5
			603		19
$AuSn \rightleftharpoons L$	692.6	$x = 0.5$	692.5	$x = 0.5$	4
$hcp \rightleftharpoons fcc + Au_5Sn$	359.3	$x^{fcc} = 0.031$ $x^{hcp} = 0.118$	333	$x^{fcc} = 0.03$ $x^{hcp} = 0.10$ (dashed points)[1]	4
$hcp \rightleftharpoons Au_5Sn + AuSn$	463.23	$x^{hcp} = 0.173$	463	$x^{hcp} = 0.185$	4
$hcp \rightleftharpoons Au_5Sn$	465.6	$x = 0.167$	468	$x = 0.167$	4
$Au_{10}Sn \rightleftharpoons fcc + hcp$	535.8	$x^{fcc} = 0.049$ $x^{hcp} = 0.111$	523	$x^{fcc} = 0.048$ $x^{hcp} = 0.11$ (dashed points)[1]	4
$L \rightleftharpoons hcp + AuSn$	555.7	$x^{L} = 0.304$ $x^{hcp} = 0.188$	551	$x^{L} = 0.29$ $x^{hcp} = 0.175$	4
			552.5		5
$Au_{10}Sn + L \rightleftharpoons hcp$	742.9	$x^{L} = 0.226$ $x^{hcp} = 0.12$	792	$x^{L} = 0.216$ $x^{hcp} = 0.138$	21
$fcc + L \rightleftharpoons Au_{10}Sn$	798	$x^{L} = 0.210$ $x^{fcc} = 0.070$	805	$x^{L} = 0.211$ $x^{fcc} = 0.071$	21
$L + fcc \rightleftharpoons hcp$	non observed		750	$x^{L} = 0.23$ $x^{fcc} = 0.068$ $x^{hcp} = 0.14$	19
$fcc + hcp \rightleftharpoons Au_{10}Sn$	non observed		730	$x^{fcc} = 0.064$ $x^{hcp} = 0.13$	19
Au(fcc) ⇌ Au(liq)	1337.58	$x = 0$	1337.58	$x = 0$	4
Sn(bct) ⇌ Sn(liq)	505.12	$x = 1$	505.12	$x = 1$	4

[1] "dashed points" means that there is no precise experimental information concerning the phase boundaries.

of invariant reaction quoted in refs. 21 and 19. From a thermodynamic point of view it means that the energetic levels of the various triphasic possibilities between these phases are very close; they can easily alter for a very small

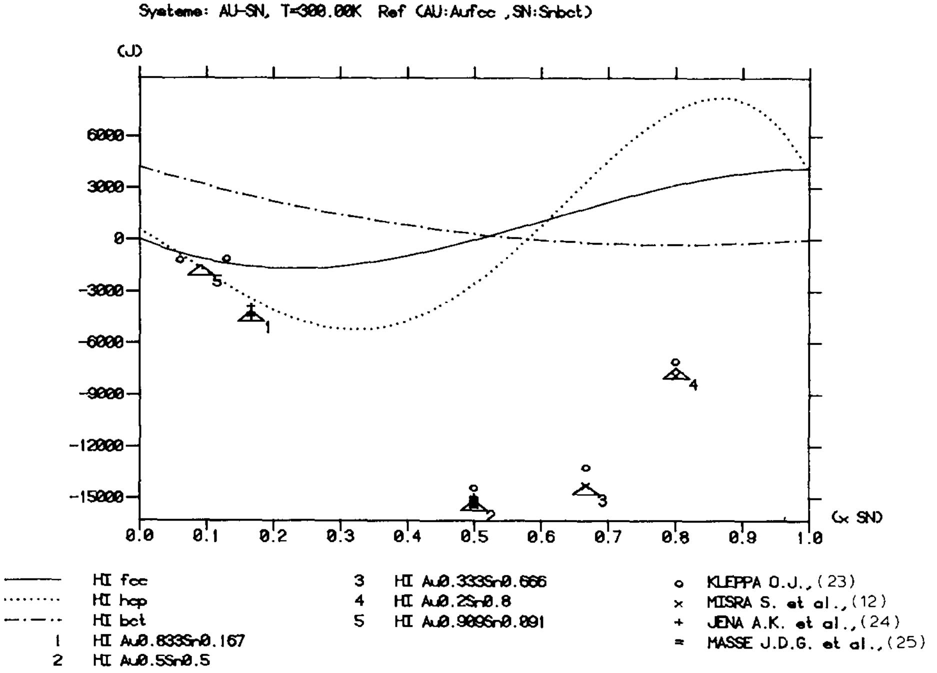

Fig. 2. Comparison between the calculated enthalpies of formation of Au–Sn solid phases and selected experimental data.

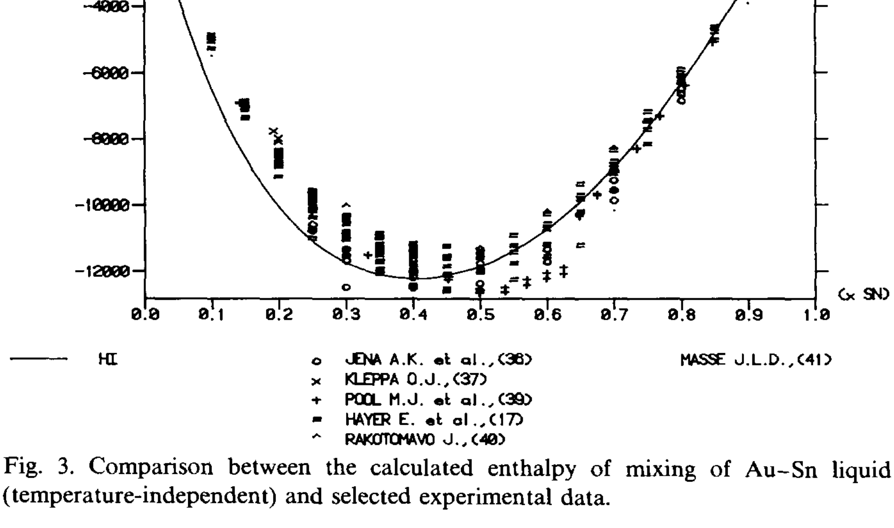

Fig. 3. Comparison between the calculated enthalpy of mixing of Au–Sn liquid alloys (temperature-independent) and selected experimental data.

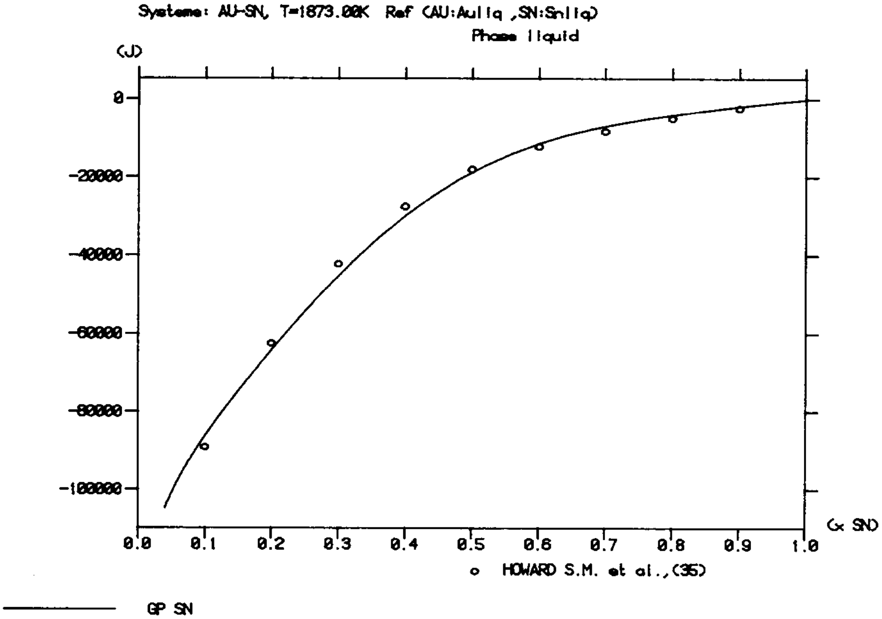

Fig. 4. Comparison between the calculated partial Gibbs energy of Sn at $T=1873$ K and experimental data of ref. 35.

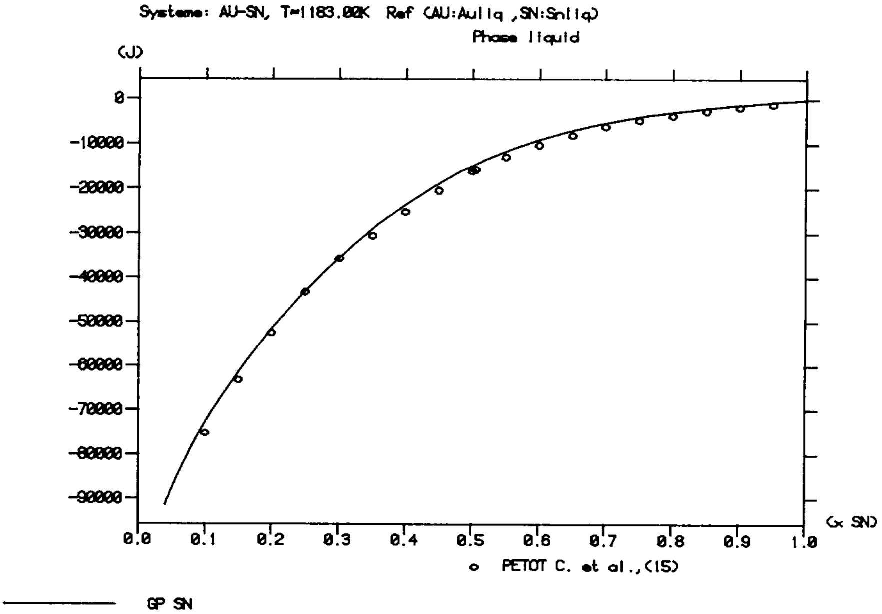

Fig. 5. Comparison between the calculated partial Gibbs energy of Sn at $T=1183$ K and experimental data of ref. 15.

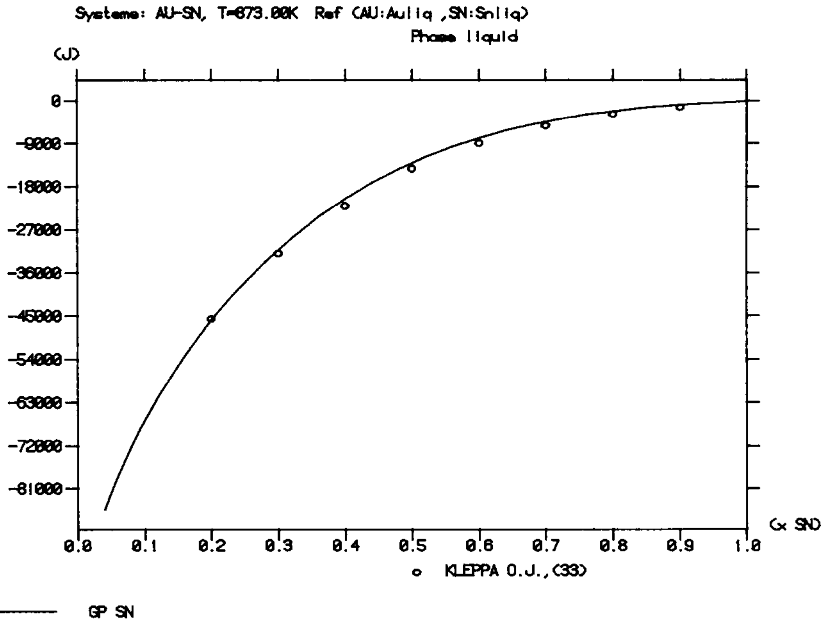

Fig. 6. Comparison between the calculated partial Gibbs energy of Sn at $T = 873$ K and experimental data of ref. 33.

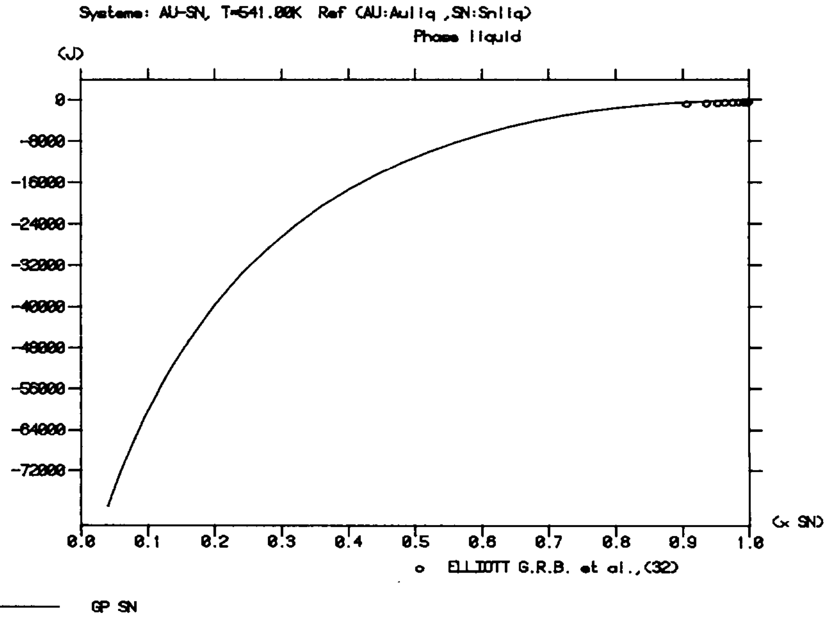

Fig. 7. Comparison between the calculated partial Gibbs energy of Sn at $T = 541$ K and experimental data of ref. 32.

change of the thermodynamic values of one of the phases, which can be obtained if new very accurate experimental measurements are available to limit more precisely the hcp phase field.

Experimental values of enthalpies of formation for the solid solution phases and stoicheiometric compounds from different authors are compared with calculated values in Fig. 2.

Figure 3 shows the good agreement obtained between the calculated enthalpy of mixing of the liquid without variation with temperature and the available experimental information resulting from calorimetry measurements. However, the agreement seems to be better in the tin-rich region than in the gold-rich region in which the calculated values are slightly more negative than the experimental ones. It may be that the thermodynamic model which was used was too simple to take into account not well understood phenomena in liquid alloys in the gold-rich region simultaneously with the experimental phase diagram, which is also very complicated in this composition range.

In Figs. 4, 5, 6, 7, the partial Gibbs energy of Sn in the liquid phase calculated at different temperatures is compared with the experimental one. The agreement is excellent.

SUMMARY

A complete thermodynamic description of the Au–Sn system is presented. Various phases of the system have been modeled and a set of self-consistent parameters has been obtained. Comparison between all the available experimental information and the calculated values seems to be very satisfactory, whereas some uncertainties remain about the invariant reaction $Au_{10}Sn + L \rightleftharpoons$ hcp in the gold rich region, which would need new experimental work.

This set of parameters is stored in the THERMODATA system, which allows us to: (i) calculate the phase diagram of the Au–Sn system; (ii) tabulate standard thermodynamic functions for various phases at any temperature, composition or reference state of the pure element; (iii) easily up-date the parameters after a new run with the optimization procedure if new experimental data are available; and (iv) predict phase equilibria in more complex multicomponent systems containing gold and tin.

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