



Thermodynamic description of the Au–Ag–Pb ternary system

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ABSTRACT

Combined with previous assessments of the Au–Ag, Ag–Pb and Au–Pb binary systems, thermodynamic description of the Au–Ag–Pb ternary system has been performed using the CALPHAD method and Thermo-calc[®] software package on the basis of the reported experimental information. The solution phases, including liquid and fcc-A1, are modeled as substitutional solutions, of which the excess Gibbs energies are expressed by the Redlich–Kister–Muggianu polynomial. The solubility of Ag in the Au–Pb intermediate compounds, Au₂Pb, AuPb₂ and AuPb₃, is not taken into account due to the lack of experimental data. Thermodynamic properties of liquid alloys, liquidus projection and several vertical sections of this ternary system have been calculated, which are in reasonable agreement with the reported experimental data. Finally, a set of self-consistent thermodynamic parameters formulating the Gibbs energies of various phases in the Au–Ag–Pb ternary system has been obtained in the present work.

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1. Introduction

Au-based alloys including Au–Sn, Au–Sb, Au–Si and Au–Ge eutectic alloys as high temperature solders are useful for bonding applications in microelectronic and optoelectronic packages [1–4]. Especially, Au–20 wt.%Sn eutectic alloy is attractive in high power electronic and optoelectronic devices because it has superior resistance to corrosion and high electrical and thermal conductivity as well as high mechanical strength [5–10]. However, to reduce the cost of Au-based solder, other elements such as Ag, Al, Bi, Cu, Ge, In, Pb, Sb, Zn, etc., may be added to replace a part of the Au. In order to understand better the role of alloying elements and to develop new Au-based solders, thermodynamic properties and phase diagrams of multi-component systems of the Au-based alloys are necessary. Recently, Many binary systems and ternary systems of the Au-based alloys such as Au–Pb, Au–Al, Au–In, Au–Zn binary systems and Au–Ag–Si, Au–Ag–Sn, Au–Bi–Sb, Au–Ge–Sn, Au–Ge–Sb, Au–Ge–Si, Au–In–Sn, Au–In–Sb, Au–Si–Sn and Au–Co–Sn ternary systems have been assessed by Wang, Liu and Jin [11–24]. Thermodynamic descriptions of the corresponding binary and ternary systems have been developed in their assessments through the CALPHAD method [25]. However, to the best of our knowledge,

no thermodynamic modelling of the Au–Ag–Pb ternary system has been reported.

The purpose of the present work is to evaluate the Au–Ag–Pb ternary system and to obtain a thermodynamic description of this ternary system using the CALPHAD method [25] and Thermo-calc[®] software package [26]. A set of self-consistent thermodynamic parameters describing various phases in this ternary system is obtained.

2. Experimental information

2.1. The sub-binary systems

The Au–Ag, Ag–Pb and Au–Pb binary systems have been assessed thermodynamically by Hassam et al. [27], Lee et al. [28] and Wang et al. [11], respectively. Good agreements are achieved between the calculated thermodynamic properties and phase diagram and experimental data. The calculated phase diagrams of the Au–Ag, Ag–Pb and Au–Pb binary systems are shown in Appendix A, respectively. The thermodynamic parameters formulating the Gibbs energies of various phases in the Au–Ag, Ag–Pb and Au–Pb binary systems obtained by Hassam et al. [27], Lee et al. [28] and Wang et al. [11] are adopted directly in the present work.

2.2. The Au–Ag–Pb ternary system

The thermodynamic properties and phase diagram of the Au–Ag–Pb ternary system were investigated experimentally by

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Table 1
Experimental information in the Au–Ag–Pb ternary system.

Reference	Method	Experimental data	Used in optimization
[29]	Electromotive force (EMF)	Activity of Pb in liquid from 1048 K to 1303 K for five constant ratios of x_{Ag}/x_{Au} Liquidus	Yes No
[30]	Calorimetry	Enthalpy of mixing of liquid at 973 K along: $Ag_{0.20}Pb_{0.80}-Au$, $Au_{0.20}Pb_{0.80}-Ag$, $Au_{0.40}Pb_{0.60}-Ag$	Yes
[31]	Differential thermal analysis	Liquidus Invariant reactions Vertical sections: 40 at.% Pb, $Au_{0.20}Ag_{0.80}-Pb$, $Au_{0.50}Ag_{0.50}-Pb$, $Au_{0.25}Pb_{0.75}-Ag$	Yes
[32]	Differential thermal analysis	Liquidus Invariant reactions Vertical sections: 40 at.% Pb, $Au_{0.20}Ag_{0.80}-Pb$, $Au_{0.50}Ag_{0.50}-Pb$, $Au_{0.25}Pb_{0.75}-Ag$	Yes

several authors [29–32]. Prince et al. [33] reviewed this ternary system and also gave recommended temperatures and compositions of the invariant reactions when compiling phase diagrams of Au-based alloys. On the basis of these experimental results in Refs. [31,32], no stable ternary compound was found in the Au–Ag–Pb ternary system. All experimental information in this ternary system are summarized in Table 1.

The thermodynamic properties of liquid alloys in the Au–Ag–Pb ternary system were investigated by Hager and Zambrano [29] and Hassam and Gheribi [30]. Using electromotive force (EMF) method, Hager and Zambrano [29] measured activities of Pb in liquid Au–Ag–Pb alloys in the temperature range of 1048–1303 K at the different sections: $x_{Ag}:x_{Au} = 5.63$, $x_{Ag}:x_{Au} = 2.30$, $x_{Ag}:x_{Au} = 1.00$, $x_{Ag}:x_{Au} = 0.18$ and $x_{Ag}:x_{Au} = 0.43$. Hassam and Gheribi [30] employed a high temperature Calvet calorimeter to determine the enthalpies of mixing of liquid Au–Ag–Pb alloys at 973 K for the following sections: $Ag_{0.20}Pb_{0.80}-Au$, $Au_{0.20}Pb_{0.80}-Ag$ and $Au_{0.40}Pb_{0.60}-Ag$. These experimental data reported by Hager and Zambrano [29] and Hassam and Gheribi [30] are used in the present optimization.

Hager and Zambrano [29] determined seven liquidus temperatures of the ternary Au–Ag–Pb alloys extrapolated from the electromotive force data. Later, through differential thermal analysis, Hassam et al. [31] determined the liquidus temperatures of the four vertical sections: $Au_{0.20}Ag_{0.80}-Pb$, $Au_{0.50}Ag_{0.50}-Pb$, $Au_{0.25}Pb_{0.75}-Ag$ and 40 at.% Pb and thus the liquidus projection was derived. Recently, Hassam and Bahari [32] investigated the phase diagram of the Au–Ag–Pb ternary system using differential thermal analysis and X-ray powder diffraction analysis. The same four vertical sections: $Au_{0.20}Ag_{0.80}-Pb$, $Au_{0.50}Ag_{0.50}-Pb$, $Au_{0.25}Pb_{0.75}-Ag$ and 40 at.% Pb were measured and two peritectic

transition reactions (U_1 : Liquid + $Au_2Pb \leftrightarrow fcc(Ag,Au) + AuPb_2$ and U_2 : Liquid + $AuPb_2 \leftrightarrow fcc(Ag,Au) + AuPb_3$) and one ternary eutectic reaction (E: Liquid $\leftrightarrow fcc(Ag,Au) + fcc(Pb) + AuPb_3$) were characterized. Experimental information reported by Hager and Zambrano [29], Hassam et al. [31] and Hassam and Bahari [32] is taken into account in the present optimization.

3. Thermodynamic modeling

3.1. Pure elements

The stable forms of the pure elements at 298.15 K and 1 bar are chosen as the reference states. The Gibbs energy for the element i in ϕ status is given as:

$${}^0G_i^\phi(T) = G_i^\phi(T) - H_i^{SER} = a + b \cdot T + c \cdot T \ln T + d \cdot T^2 + e \cdot T^3 + f \cdot T^{-1} + g \cdot T^7 + h \cdot T^{-9} \quad (1)$$

where H_i^{SER} is the enthalpy of the element i in its standard reference state (SER) at 298.15 K and 1 bar; T is the absolute temperature in K; $G_i^\phi(T)$ is the Gibbs energy of the element i with structure ϕ . In the present work, the Gibbs energies of the elements Ag, Au and Pb, ${}^0G_{Ag}^\phi(T)$, ${}^0G_{Au}^\phi(T)$ and ${}^0G_{Pb}^\phi(T)$ are taken from the SGTE (Scientific Group Thermodata Europe) database compiled by Dinsdale [34].

3.2. Solution phases

The substitutional solution model is employed to describe the solution phases including liquid and fcc.A1(Au), respectively. The molar Gibbs energy of the solution phase ϕ (ϕ = liquid, and fcc.A1)

Table 2
Thermodynamic parameters for the Au–Ag–Pb ternary system.

Phase	Thermodynamic parameter ^a	Reference
Liquid (Ag,Au,Pb)	${}^{(0)}L_{Ag,Au} = -16,042 + 1.14T$	[27]
	${}^{(0)}L_{Ag,Pb} = +13,330.32 - 6.2127T$	
	${}^{(1)}L_{Ag,Pb} = -1449.03 - 1.05718T$	[28]
	${}^{(2)}L_{Ag,Pb} = -2089.13$	
	${}^{(0)}L_{Au,Pb} = -17,577.988 + 105.97303T - 14.435179T \ln T$	[11]
	${}^{(1)}L_{Au,Pb} = +2673.2826 - 5.379358T$	
	${}^{(0)}L_{Ag,Au,Pb} = +6000 - 5T$	
fcc.A1 (Ag,Au,Pb)	${}^{(1)}L_{Ag,Au,Pb} = +24,000 - 23T$	This work
	${}^{(2)}L_{Ag,Au,Pb} = -5000 + 5T$	
	${}^{(0)}L_{Ag,Au} = -15,599$	[27]
	${}^{(0)}L_{Ag,Pb} = +30,325.21 - 5.74345T$	[28]
Au ₂ Pb (Au) _{0.667} (Pb) _{0.333}	${}^{(1)}L_{Ag,Pb} = -3290.47$	[11]
	${}^{(0)}L_{Au,Pb} = +30,000$	
	$G_{Au_2Pb}^{Au,Pb} = 0.667{}^0G_{Au}^{fcc} + 0.333{}^0G_{Pb}^{fcc} - 3010 + 0.5T$	[11]
AuPb ₂ (Au) _{0.333} (Pb) _{0.667}	$G_{AuPb_2}^{Au,Pb} = 0.333{}^0G_{Au}^{fcc} + 0.667{}^0G_{Pb}^{fcc} - 2800 + 1.45T$	[11]
AuPb ₃ (Au) _{0.25} (Pb) _{0.75}	$G_{AuPb_3}^{Au,Pb} = 0.25{}^0G_{Au}^{fcc} + 0.75{}^0G_{Pb}^{fcc} - 1900 + 0.56T$	[11]

^a Note: Gibbs energies are expressed in J/mol. The lattice stabilities of the elements Au, Ag and Pb in liquid and fcc.A1 were given by Dinsdale [34].

Table 3
Invariant reactions in the Au–Ag–Pb ternary system.

System	Reaction	Type	T (K)	Composition		Reference
				x_{Ag}^L	x_{Au}^L	
Au–Pb system	$L + fcc(Au) \leftrightarrow Au_2Pb$	p ₁	707	–	0.640	[11]
	$L + Au_2Pb \leftrightarrow AuPb_2$	p ₂	526	–	0.259	
	$L + AuPb_2 \leftrightarrow AuPb_3$	p ₃	495	–	0.174	
	$L \leftrightarrow AuPb_3 + fcc(Pb)$	e ₁	488	–	0.152	
Ag–Pb system	$L \leftrightarrow fcc(Ag) + fcc(Pb)$	e ₂	577	0.043	–	[28]
Au–Ag–Pb system	$L + Au_2Pb \leftrightarrow fcc(Ag,Au) + AuPb_2$	U ₁	524	0.0053	0.2610	[31]
			491	–	–	[32]
			522	–	–	[33]
			524	0.0043	0.2596	This work
	$L + AuPb_2 \leftrightarrow fcc(Ag,Au) + AuPb_3$	U ₂	496	0.0048	0.1861	[31]
			487	–	–	[32]
			491	–	–	[33]
			493	0.0051	0.1738	This work
	$L \leftrightarrow fcc(Ag,Au) + fcc(Pb) + AuPb_3$	E	487	0.0051	0.1696	[31]
			482.5	–	–	[32]
485			–	–	[33]	
486			0.0052	0.1521	This work	

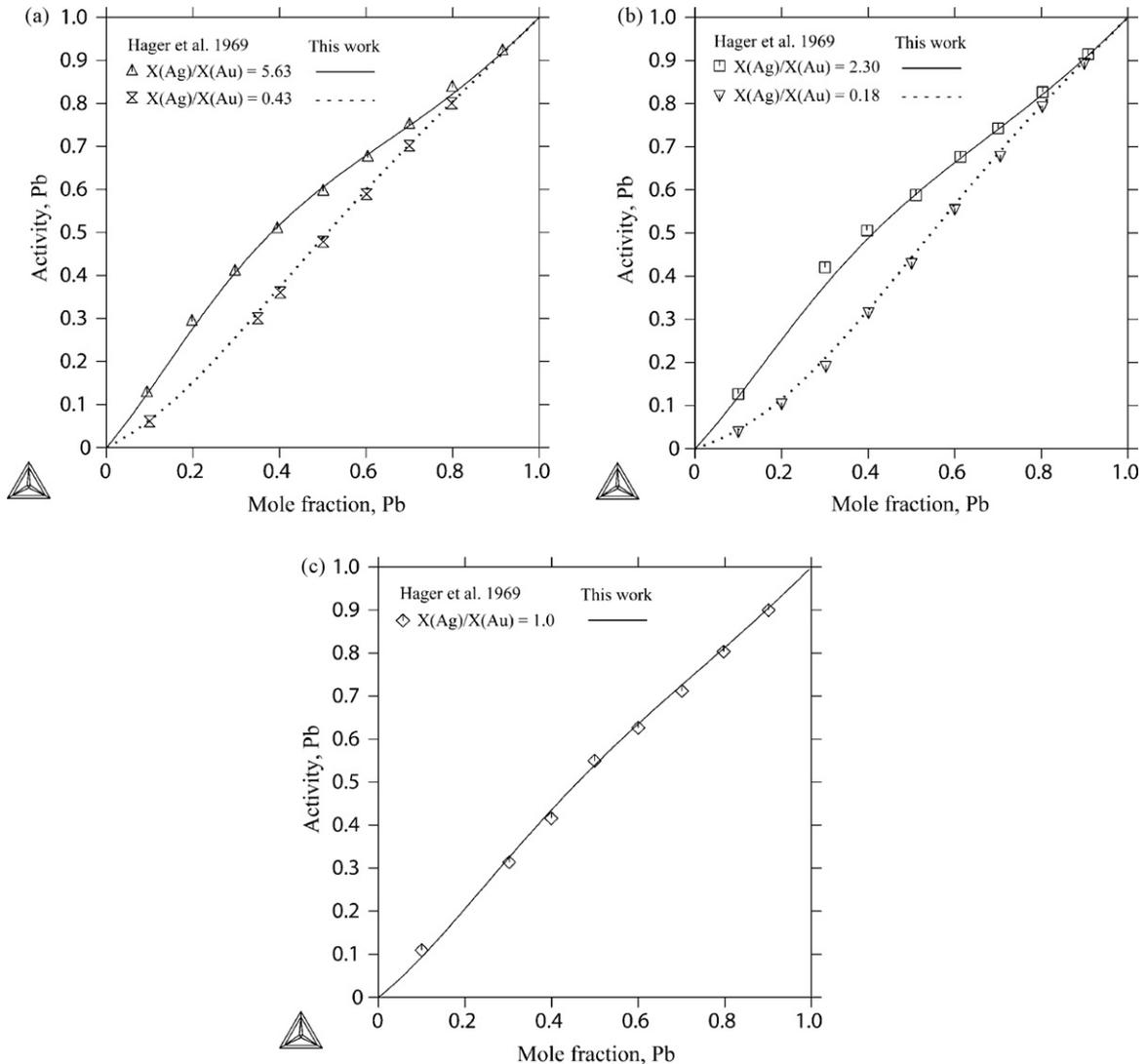


Fig. 1. Comparison of the calculated activities of Pb in liquid ternary alloys with experimental data [29] referred to liquid Pb at 1200 K. (a) $x_{Ag}:x_{Au} = 5.63$ and $x_{Ag}:x_{Au} = 0.43$; (b) $x_{Ag}:x_{Au} = 2.30$ and $x_{Ag}:x_{Au} = 0.18$; (c) $x_{Ag}:x_{Au} = 1.0$.

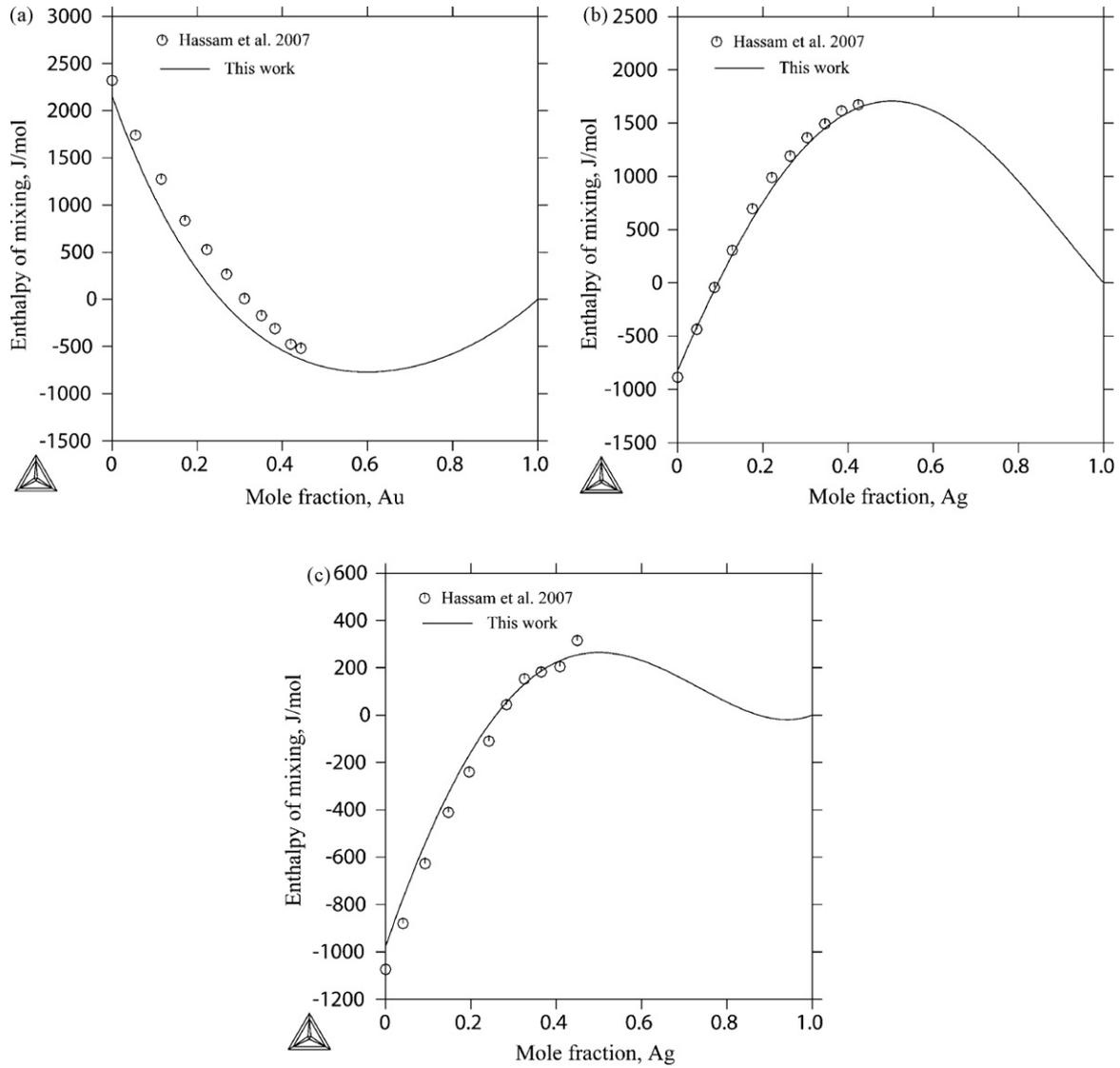


Fig. 2. Comparison of the calculated enthalpies of mixing of liquid ternary alloys with experimental data [30] referred to liquid Ag, Au and Pb at 973 K. (a) $\text{Ag}_{0.20}\text{Pb}_{0.80}\text{-Au}$; (b) $\text{Au}_{0.20}\text{Pb}_{0.80}\text{-Ag}$ and (c) $\text{Au}_{0.40}\text{Pb}_{0.60}\text{-Ag}$.

can be expressed as:

$$G_m^\phi = \sum x_i {}^0G_i^\phi + RT \sum x_i \ln(x_i) + {}^E G_m^\phi \quad (2)$$

where ${}^0G_i^\phi$ is the molar Gibbs energy of the element i ($i = \text{Ag}, \text{Au}, \text{Pb}$) with the structure ϕ , x_i the mole fraction of component i , R gas constant, T temperature in K, ${}^E G_m^\phi$ the excess Gibbs energy. The excess Gibbs energy of phase ϕ can be expressed by the Redlich–Kister polynomial [35] as:

$${}^E G_m^\phi = x_{\text{Ag}}x_{\text{Au}} \sum_{j=0}^n {}^{(j)}L_{\text{Ag,Au}}^\phi (x_{\text{Ag}} - x_{\text{Au}})^j + x_{\text{Ag}}x_{\text{Pb}} \sum_{j=0}^n {}^{(j)}L_{\text{Ag,Pb}}^\phi (x_{\text{Ag}} - x_{\text{Pb}})^j + x_{\text{Au}}x_{\text{Pb}} \sum_{j=0}^n {}^{(j)}L_{\text{Au,Pb}}^\phi (x_{\text{Au}} - x_{\text{Pb}})^j + x_{\text{Ag}}x_{\text{Au}}x_{\text{Pb}}L_{\text{Ag,Au,Pb}}^\phi \quad (3)$$

where ${}^{(j)}L_{\text{Ag,Au}}^\phi$, ${}^{(j)}L_{\text{Ag,Pb}}^\phi$ and ${}^{(j)}L_{\text{Au,Pb}}^\phi$ are binary interaction parameters, which are taken directly from the Au–Ag, Ag–Pb and Au–Pb systems assessed by Hassam et al. [27], Lee et al. [28] and Wang et al. [11], respectively. The ternary interaction parameter $L_{\text{Ag,Au,Pb}}^\phi$ is formulated with Redlich–Kister–Muggianu expression [36] as

follows:

$$L_{\text{Ag,Au,Pb}}^\phi = x_{\text{Ag}}{}^{(0)}L_{\text{Ag,Au,Pb}} + x_{\text{Au}}{}^{(1)}L_{\text{Ag,Au,Pb}} + x_{\text{Pb}}{}^{(2)}L_{\text{Ag,Au,Pb}} \quad (4)$$

where ${}^{(j)}L_{\text{Ag,Au,Pb}}$ are parameters to be evaluated in the present work.

3.3. Intermetallic compounds

In the Au–Ag–Pb ternary system, the solubilities of Ag in Au_2Pb , AuPb_2 and AuPb_3 are not considered due to the lack of the available experimental information. Thus the Gibbs energies of the binary intermetallic compounds Au_2Pb , AuPb_2 and AuPb_3 are taken directly from the Au–Pb binary system optimized by Wang et al. [11] during the present optimization and calculation of this ternary system.

4. Results and discussion

Using the compatible lattice stabilities of the elements Ag, Au and Pb compiled by Dinsdale [34], the model parameters for liquid phase in the Au–Ag–Pb ternary system are optimized using the PARROT module in the Thermo-calc[®] software package developed

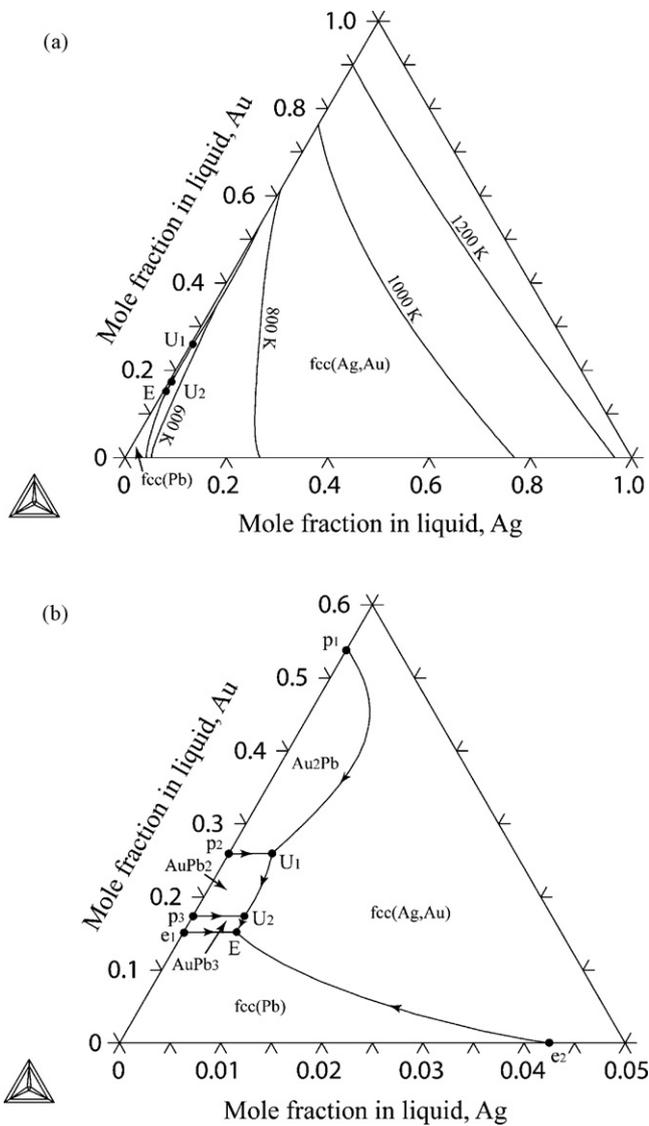


Fig. 3. Calculated liquidus projection of the Au–Ag–Pb ternary system in the present work. (a) Overview and (b) Pb-rich part.

by Sundman et al. [26]. This module works through minimizing the square sum of the differences between experimental data and calculated values. During the present optimization procedure, each set of experimental data is given a certain weight according to the reliability and compatibility of experimental data. It should be claimed that the weights of much more reliable experimental data are higher than that of less ones during the optimization. For thermodynamic data, in general, the activity of a component measured by the EMF method are much more reliable and within a smaller experimental error than that determined by vapour pressure method, while the enthalpy of mixing of liquid alloys obtained directly by calorimetry are much more believable than that derived from indirect measurements by other methods (such as EMF). As for phase diagram data, it is better to use the measured experimental data by thermal analysis method in the optimization.

Thermodynamic parameters for all condensed phases in the Au–Ag–Pb system are summarized in Table 2. Table 3 shows invariant reactions for Ag–Pb and Au–Pb binary systems and the Ag–Au–Pb ternary system. Thermodynamic properties of liquid alloys, liquidus projection and several vertical sections of this ternary system are also calculated and compared with the experimental data as illustrated in Figs. 1–7. Reasonable agreements

Table 4
Liquidus temperatures of ternary Au–Ag–Pb alloys.

Alloy composition			Liquidus temperature, T_{liq} (K)	
x_{Ag}	x_{Au}	x_{Pb}	Experiment [29]	Calculation
0.7693	0.1367	0.094	1163	1157
0.6810	0.1210	0.198	1061	1051
0.6273	0.2727	0.100	1182	1168
0.4500	0.4500	0.100	1175	1186
0.2706	0.6294	0.100	1219	1199
0.1373	0.7627	0.100	1228	1204
0.1220	0.6780	0.200	1101	1080

are achieved between the calculated results and the experimental data.

Fig. 1 shows comparisons between the calculated activities of Pb in liquid Au–Ag–Pb ternary alloys and the experimental data measured by Hager and Zambrano [29] at 1200 K. As can be seen, the calculated activities of Pb in liquid phase along five cross sections with Ag to Au ratio of 5.63, 2.30, 1.00, 0.18 and 0.43 at 1200 K are in excellent agreement with the experimental data [29].

Fig. 2 compares the calculated enthalpies of mixing of liquid Au–Ag–Pb ternary alloys with the experimental data reported by Hassam and Gheribi [30] at 973 K. The calculated enthalpies of mixing of liquid phase for the cross sections (Au_{0.20}Pb_{0.80}–Ag and Au_{0.40}Pb_{0.60}–Ag) agree well with experimental data [30] as given in Fig. 2(b) and (c). Fig. 2(a) also presents a slight discrepancy between the calculated enthalpies of mixing of liquid alloys along the cross section (Ag_{0.20}Pb_{0.80}–Au) and the experimental data measured by Hassam and Gheribi [30]. From Fig. 2(a), it is concluded that the calculated enthalpy of mixing of liquid in the Ag–Pb binary system in Ref. [28] is much negative than experimental data [30]. Thus, this discrepancy originates mainly from enthalpy of mixing of the Ag–Pb binary system. Furthermore, the derivation is generally within experimental error because the relative experimental error of enthalpies of mixing of liquid alloys was given to be about 5% in Ref. [30].

In the Au–Ag–Pb ternary system, all invariant reactions (U_1 , U_2 and E) are also given in Fig. 3 and Table 3. From Table 3, the calculated temperatures of invariant reactions (U_1 at 524 K and U_2 at 493 K) agree well with the experimental data given by Hassam et al. [31] and the values assessed by Prince et al. [33], but deviate from the experimental data (U_1 at 491 K and U_2 at 487 K) measured

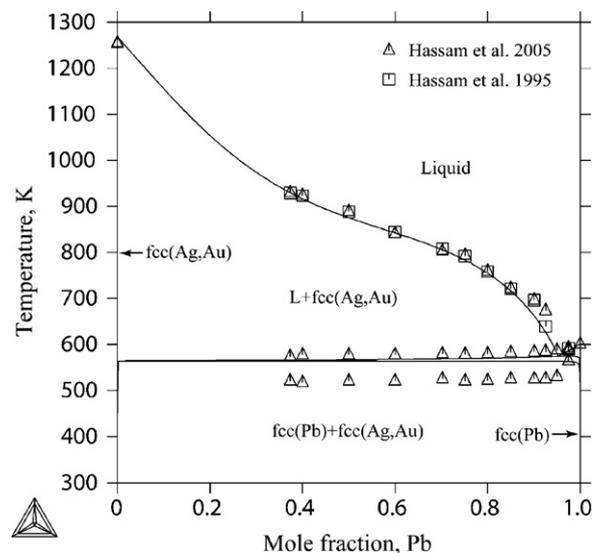


Fig. 4. Calculated vertical section of Au_{0.20}Ag_{0.80}–Pb and compared with experimental data [31,32].

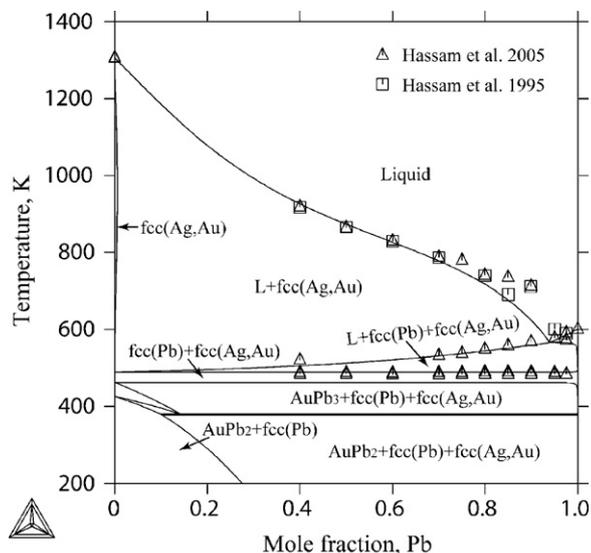


Fig. 5. Calculated vertical section of $\text{Au}_{0.50}\text{Ag}_{0.50}\text{-Pb}$ and compared with experimental data [31,32].

by Hassam and Bahari [32], respectively. This deviation is mainly originated from experimental error because the temperatures of all invariant reactions in this ternary system are very close to these of invariant reactions in the Au–Pb binary system. It is difficult to measure accurately reaction temperatures because of the small temperature range. On the other hand, according to the experimental conditions, the experimental results obtained by Hassam et al. [31] are more reliable than the measured values in Ref. [32] because the alloy samples were annealed for six months at 473 K before thermal analysis in order to decrease thermal effect at low temperature. In addition, care and great attention were paid to fix well the latest experimental data measured by Hassam and Bahari [32] during the present optimization. However, this attempt was not successful. Meanwhile, the calculated temperature of eutectic reaction E (486 K) is in good agreement with the experimental data reported by Hassam et al. [31], Hassam and Bahari [32] and the values assessed by Prince et al. [33]. As shown in Table 3, the differences between the calculated equilibrium compositions of liquid

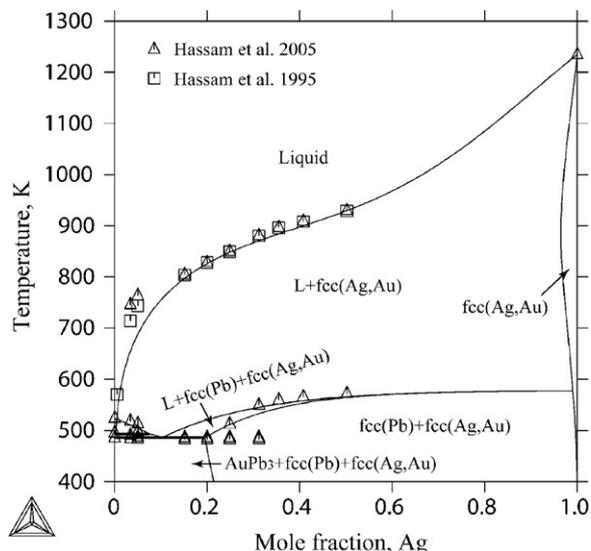


Fig. 6. Calculated vertical section of $\text{Au}_{0.25}\text{Pb}_{0.75}\text{-Ag}$ and compared with experimental data [31,32].

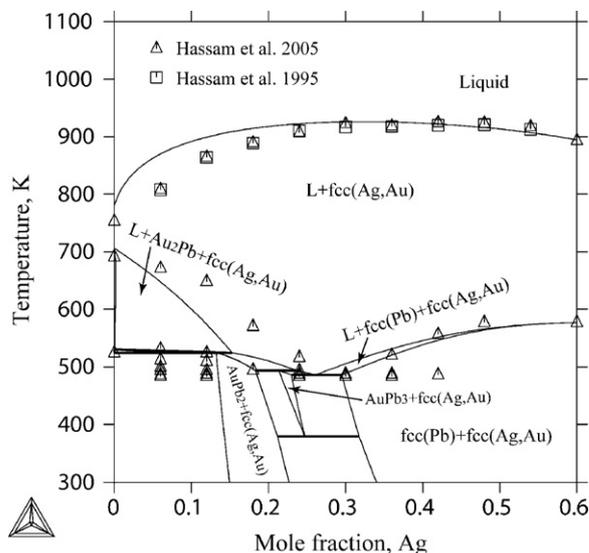


Fig. 7. Calculated vertical section at 40 at.% Pb and compared with experimental data [31,32].

phase for all invariant reactions (U_1 , U_2 and E) and the experimental data measured by Hassam et al. [31] are very small (about 2 at.%).

Table 4 compares the calculated liquidus temperatures of seven alloys with the experimental results extrapolated from the electromotive force data by Hager and Zambrano [29]. There is an obvious difference between the calculated liquidus temperatures and experimental values [29]. It should be pointed out that large error may exist in such deduced values of liquidus temperatures in comparison with the data obtained directly using thermal analysis.

Figs. 4–7 present the calculated vertical sections of $\text{Au}_{0.20}\text{Ag}_{0.80}\text{-Pb}$, $\text{Au}_{0.50}\text{Ag}_{0.50}\text{-Pb}$, $\text{Au}_{0.25}\text{Pb}_{0.75}\text{-Ag}$ and 40 at.% Pb with the experimental data determined by Hassam et al. [31], Hassam and Bahari [32]. As can be seen, the calculated phase relations and phase boundaries of liquid phase in Figs. 4–6 are in good agreement with experimental data [31,32]. In Fig. 7, the calculated phase relations of the vertical section at 40 at.% Pb is consistent with the experimental results, while the calculated phase boundaries of liquid phase in the Au–Pb side show a slight deviation from the experimental data measured by Hassam et al. [31], Hassam and Bahari [32].

5. Conclusions

Based on the previous assessments of the Au–Ag, Ag–Pb and Au–Pb binary systems and available experimental information on the Au–Ag–Pb ternary system, the thermodynamic description of the Ag–Au–Pb ternary system has been developed using the CALPHAD method and Thermo-calc[®] software package. A set of self-consistent thermodynamic parameters formulating the Gibbs energies of various phases in the Au–Ag–Pb ternary system have been obtained in the present work, which can be used to reproduce most experimental data including thermodynamic properties and phase equilibria data.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.tca.2010.04.003.

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