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Thermodynamic assessment of the Ca–Pb system

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Abstract

Thermodynamic modelling of the Ca–Pb system was carried out. The thermodynamic parameters were determined using available experimental information: phase diagram data and thermodynamic quantities. For this system, the liquid phase was described from the association model with the "Ca_2Pb" associated complex. The calculations well restore experimental values. All the results so-obtained (optimised thermodynamic parameters and calculated phase diagram) are reported in this paper and discussed.

Keywords: Calcium–lead system; Thermodynamic modelling; Association model

1. Introduction

The Ca–Pb and Ca–Pb–Sn alloys are studied for many years for their industrial application as electrode material in lead acid batteries. For the Ca–Pb alloys, the introduction of calcium in lead leads to a structural hardening after quenching by precipitation of intermetallic compound. The mechanical and electrochemical behaviour of the Ca–Pb alloys is influenced by their metallurgical state. In consequence, the knowledge of phase boundaries in the Ca–Pb system will be crucial for predicting and increasing metallurgical behaviour in the electrode materials. It will permit us to explain the evolution of age-hardening mechanism in the Ca–Pb and Ca–Pb–Sn systems [1].

The purpose of this study is the thermodynamic modelling of the Ca–Pb system, the first step of the modelling of the ternary Ca–Pb–Sn system, using the CALPHAD (CALculation of Phase Diagram) method and Thermocalc^{TM} software.

2. Bibliographic review

2.1. Phase diagram information

The calcium–lead system was investigated by many workers. Itkin and Alcock made an exhaustive review [2–22] and proposed the phase diagram presented in Fig. 1, in 1992 [2]. It is mainly based on the thermal analyses of Bruzzone and Merlo [3] and can be described as follows:

- Four intermediate compounds exist (Table 1): Ca_2Pb [3–5] and $CaPb_3$ [3,6–8] characterised by congruently melting, and CaPb [3,4,7] and Ca_5Pb_3 [3,9] formed by peritectic reaction.
- αCa and βCa solid solutions assumed to dissolve negligible amounts of Pb.
- The value of calcium solubility in the primary lead-solid solution is not clearly defined at the invariant equilibrium (599.35 K). It depends on the kind of the invariant equilibrium (eutectic or peritectic). This fact has been discussed by several authors: Vigdorovich and Nashel'skii [10] assumed the existence of an eutectic reaction from DTA and microhardness measurements. It must be noted that this latter conclusion was obtained by the extrapolation of the solvus line. Contrarily, Shumacher and Bouton [11] found by resistivity and hardness measurements a peritectic reaction at 601.15 K. Recently, Notin et al. [12] confirmed the result given in Ref. [10]. They compared the thermal behaviour of pure lead and of lead alloys weakly enriched in calcium by DSC. It appeared that the invariant reaction temperature in this domain of composition is about 1 K higher than the lead melting temperature. The apparatus used in this study was a Calvet microcalorimeter well known for its good sensitivity.

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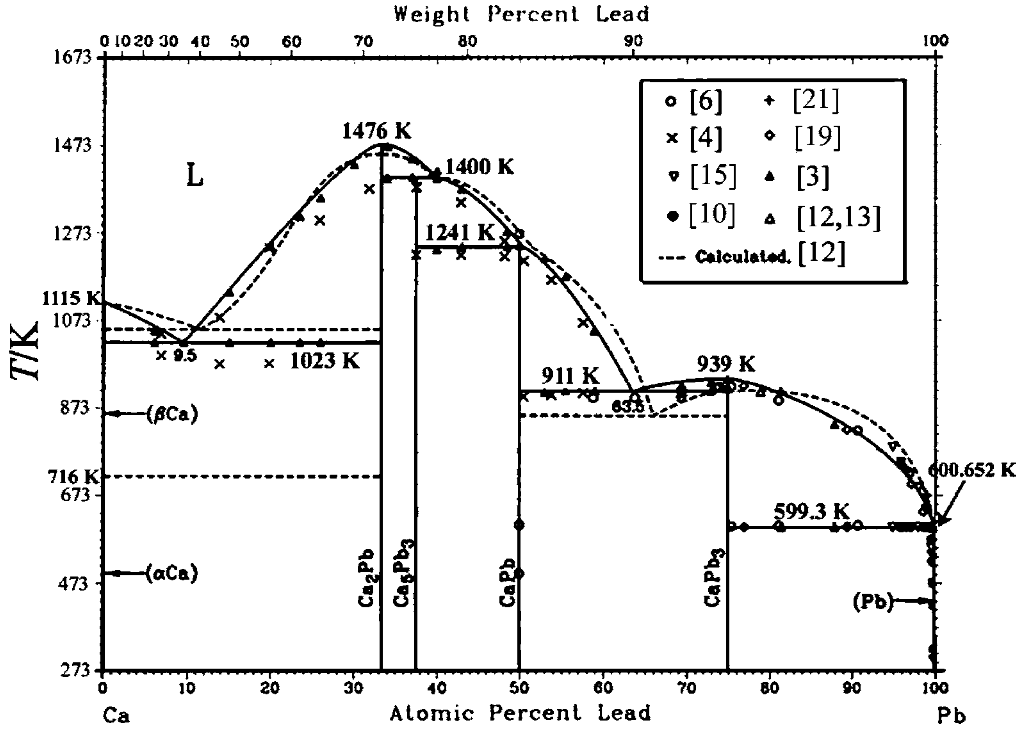

Fig. 1. Ca–Pb phase diagram furnished by Itkin and Alcock [2].

Table 1
Crystallographic structure for the intermediate phases of the Ca–Pb system

Phase	Composition (at.% Pb)	Pearson symbol	Space group	Strukturbericht designation	Prototype	Reference
Ca_2Pb	33.3	*oP*12	*Pnma*	*C*23	Co_2Si	[5]
Ca_5Pb_3	37.5	*hP*48	$P6_3mc$	–	Ca_5Pb_3	[9]
CaPb	50	*tP*4	*P*4/*mmm*	$L1_0$	AuCu	[3]
$CaPb_3$	75	*cP*4	$Pm\bar{3}m$	$L1_2$	$AuCu_3$	[8]

- Bouirden and coworkers [12,13] measured the liquidus temperature of alloys for compositions close to that of $CaPb_3$ by calorimetry. The same authors have performed a calculation of the Ca–Pb phase diagram.

2.2. Thermodynamic data

Several thermodynamic information are available in the literature concerning the Ca–Pb system [12–20]. Notin et al. [12] and Bouirden [13] measured by drop calorimetry at 910 and 1042 K the integral enthalpy of mixing as a function of calcium content ($0 < x(\mathrm{Ca}) < 0.5$). The results are presented in Figs. 2 and 3. The variation of the integral enthalpy of mixing in the liquid phase versus calcium molar fraction is linear which indicates the presence of an associated complex [23]. Notin et al. [12] and Bouirden [13] determined the enthalpy of formation of the intermetallic compounds ($CaPb_3$ and CaPb) (Table 2) by dissolution calorimetry in a pure lead-bath.

Several authors measured by an e.m.f. method the calcium activity for different composition ranges in Ca–Pb system. Delcet et al. [16] gave information (using a CaF_2 electrolyte) for liquid alloys containing more than 60 at.% lead at 1073 K. Nouri et al. [17] measured (using a CaF_2 electrolyte) as a function of temperature between 703 and 883 K in the Pb–Pb_3Ca two-phase domain. Finally, Fray and Kumar [18]

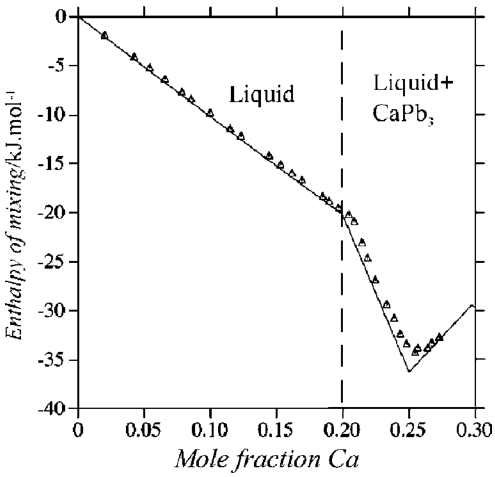

Fig. 2. Calculated integral mixing enthalpy in Ca–Pb system at 910 K compared with data from Bouirden [13] (reference state: $Ca_{(s)}$; $Pb_{(l)}$).

Table 2
Comparison between experimental and calculated values of enthalpy of formation of Ca–Pb intermetallic compounds

Intermetallic compounds	Reference state	Experimental value determined by Bouirden [13] (kJ mol^{-1})	T (K)	Calculated value (kJ mol^{-1})	Relative discrepancy (%)
$CaPb_3$	Ca(β); Pb(l)	−35.0 ± 0.3	900	−36.251	3.6
CaPb	Ca(β); Pb(l)	−57 ± 2	1040	−58.664	2.9
Ca_5Pb_3	Ca(α); Pb(α)	−56 ± 1	300	−69.354	23.8
Ca_2Pb	Ca(α); Pb(α)	−59.0 ± 1.5	300	−69.577	17.9

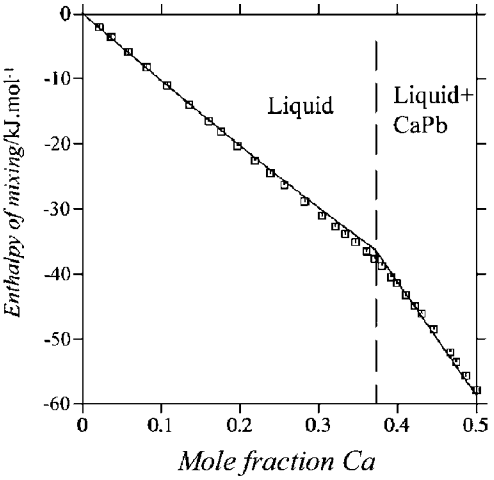

Fig. 3. Calculated integral mixing enthalpy in Ca–Pb system at 1042 K compared with data from Bouirden [13] (reference state: $Ca_{(s)}$; $Pb_{(l)}$).

reported calcium activity values at 1173 K (using a calcium magnetoplumbite electrolyte) for calcium contents from 0 to 70 at.%.

3. Thermodynamic modelling

3.1. Pure elements

The Gibbs energy of pure elements i with the structure ϕ, referred to the enthalpy of its standard state element reference (SER) at 298.15 K, H_i^{SER}, is described as a function of the temperature by the following equation:

$$ {}^{0}G_i^{\phi}(T) - H_i^{\mathrm{SER}} = a + bT + cT\ln T + dT^2 + eT^3 + fT^{-1} + gT^7 + hT^{-9} \quad (1) $$

This quantity is denoted GHSERi when the structure ϕ corresponds to SER. Values of the coefficients, a, b,..., h, are taken from the SGTE database [24].

3.2. Primary solid solution

The solid face-centred cubic (FCC)_A1(Pb) and (FCC)_A1(Ca), and body-centred cubic (BCC)_A2(Ca) solid solutions are described by a random substitutional model and their molar Gibbs energy are given by the general formula:

$$ G^{\phi} = {}^{\mathrm{ref}}G^{\phi} + {}^{\mathrm{id}}G^{\phi} + {}^{\mathrm{ex}}G^{\phi} \quad (2) $$

where ${}^{\mathrm{ref}}G^{\phi}$ is the contribution of the pure components of the phase to the Gibbs energy, ${}^{\mathrm{id}}G^{\phi}$ the ideal mixing contribution and ${}^{\mathrm{ex}}G^{\phi}$ the excess Gibbs energy corresponding to the non-ideal interactions between the components.

These three terms are expressed by the following equations:

$$ {}^{\mathrm{ref}}G^{\phi} = \sum_i x_i {}^{0}G_i^{\phi} \quad (3) $$

$$ {}^{\mathrm{id}}G^{\phi} = RT\sum_i x_i \ln x_i \quad (4) $$

$$ {}^{\mathrm{ex}}G^{\phi} = x_i x_j \sum_{\nu=0} {}^{\nu}L_{i,j}^{\phi}(x_i - x_j)^{\nu} \quad (5) $$

where x_i represents the molar fraction of component i and ${}^{\nu}L_{i,j}^{\phi}$ a binary interaction parameter described with a linear dependence:

$$ {}^{\nu}L_{i,j}^{\phi} = a_{\nu} + b_{\nu}T \quad (6) $$

3.3. Liquid phase

The liquid phase was described using the association model [23]. The Gibbs energy is given for 1 mol of formula units and is expressed as the sum of four contributions:

$$ G^{\mathrm{liq}} - H^{\mathrm{SER}} = {}^{\mathrm{ref}}G + {}^{\mathrm{form}}G + {}^{\mathrm{id}}G^{\mathrm{liq}} + {}^{\mathrm{ex}}G^{\mathrm{liq}} \quad (7) $$

where the different contributions are the reference part (${}^{\mathrm{ref}}G$), formation of the associated complex part (${}^{\mathrm{form}}G$), an ideal part (${}^{\mathrm{id}}G^{\mathrm{liq}}$) and an excess part (${}^{\mathrm{ex}}G^{\mathrm{liq}}$).

The association model is based on the hypothesis that a complex C is formed in the liquid phase:

$$ pA + qB \leftrightarrow A_pB_q \ (A_pB_q \text{ corresponds to the complex C}) \quad (8) $$

where A and B are pure elements initially present in the mixing, and p and q are the stoichiometric coefficients. Thus, the different parts of the Gibbs energy can be described by Eqs. (9)–(11):

$$ {}^{\mathrm{ref}}G = \{x_A[{}^{0}G_A^{\mathrm{liq}}(T) - H_A^{\mathrm{SER}}] + x_B[{}^{0}G_B^{\mathrm{liq}}(T) - H_B^{\mathrm{SER}}]\}(y_A + (p+q)y_C + y_B) \quad (9) $$

$$ {}^{\mathrm{form}}G = (y_A {}^{0}G_A^{\mathrm{liq}}) + (y_B {}^{0}G_B^{\mathrm{liq}}) + (y_C {}^{0}G_C^{\mathrm{liq}}) \quad (10) $$

Table 3
Calculated and experimental (values in parentheses) temperatures and compositions of invariant reactions in the Ca–Pb system

Reaction	Composition of the respective phases (at.% Pb)			T (K)	Type of equilibrium	Reference
Liq ↔ βCa		0		1115.00	Melting	–
βCa ↔ αCa		0 (*0*)		716.03 (*716.15*)	Allotropic	[26]
Liq ↔ βCa + Ca_2Pb	9.74 (*9.50*)	0 (*0*)	33.3 (*33.3*)	1023.13 (*1023.15*)	Eutectic	[3]
Liq ↔ Ca_2Pb		33.3 (*33.3*)		1476.52 (*1476.15*)	Congruent	[3]
Liq + Ca_2Pb ↔ Ca_5Pb_3	40.78 (*∼40.00*)	33.3 (*33.3*)	37.5 (*37.5*)	1403.23 (*1400.15*)	Peritectic	[3]
Liq + Ca_5Pb_3 ↔ CaPb	50.02 (*∼50.50*)	37.5 (*37.5*)	50 (*50*)	1238.04 (*1241.15*)	Peritectic	[3]
Liq ↔ CaPb + $CaPb_3$	68.52 (*∼63.50*)	50 (*50*)	75 (*75*)	896.79 (*911.15*)	Eutectic	[3]
Liq ↔ $CaPb_3$		75 (*75*)		924.21 (*939.15*)	Melting	[3]
Liq ↔ $CaPb_3$ + Pb	99.60 (*∼99.64*)	75 (*75*)	99.61 (*∼99.49*)	602.52 (*601.90*)	Peritectic (*Peritectic*)	[10,11]
Liq ↔ Pb		1		600.65	Melting	–

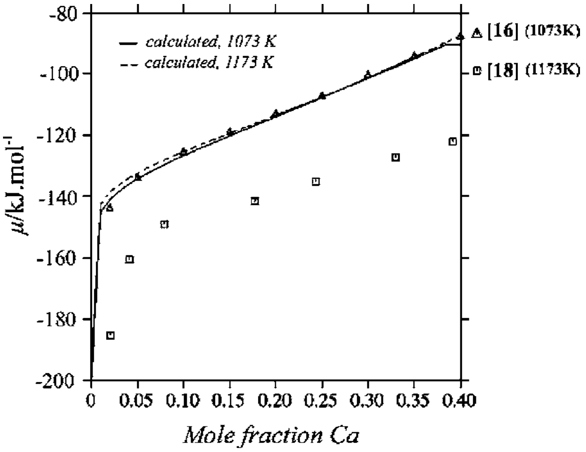

Fig. 4. Comparison between calculated and measured calcium activity by Delcet et al. [16] at 1073 K and Fray and Kumar [18] at 1173 K (reference state: $Ca_{(l)}$).

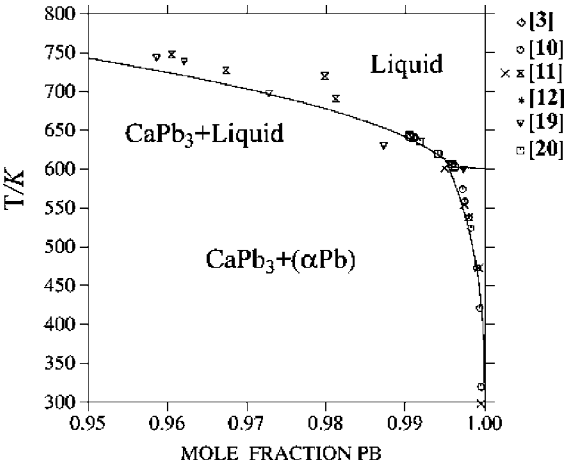

Fig. 6. Ca–Pb calculated phase diagram and data issued from literature in the lead-rich corner.

$$
{}^{\mathrm{id}}G^{\mathrm{liq}} = RT(y_{\mathrm{A}}\ln y_{\mathrm{A}} + y_{\mathrm{B}}\ln y_{\mathrm{B}} + y_{\mathrm{C}}\ln y_{\mathrm{C}}) \tag{11}
$$

where R is the perfect gas constant, T the temperature; A, B and C are the three species in the liquid and y_{A}, y_{B} and y_{C} their molar fractions normalised to $y_{\mathrm{A}} + y_{\mathrm{B}} + y_{\mathrm{C}} = 1$, respectively. x_{A} and x_{B} are the initial molar fractions introduced in the solution, without considering the associated species, normalised to $x_{\mathrm{A}} + x_{\mathrm{B}} = 1$.

The excess Gibbs energy is expressed by the Redlich-Kister polynomial:

$$
{}^{\mathrm{ex}}G^{\mathrm{liq}} = y_i y_j \sum_{\nu=0} {}^{\nu}L^{\mathrm{liq}}_{i,j}(y_i - y_j)^{\nu} \tag{12}
$$

where i and j are indexes which correspond to the A, B or C species. The binary interaction parameter is described as a function of temperature by Eq. (6).

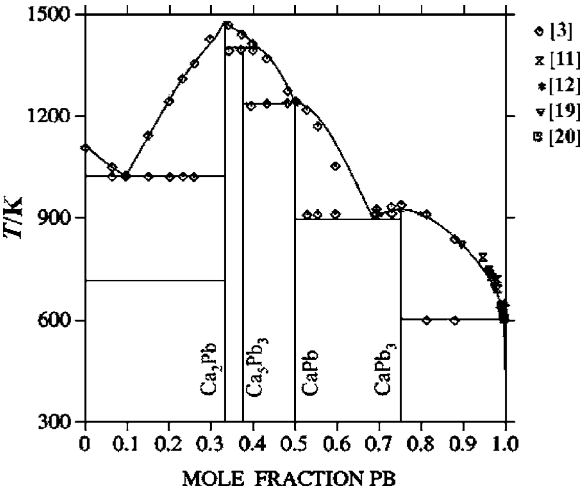

Fig. 5. Ca–Pb calculated phase diagram and data issued from literature.

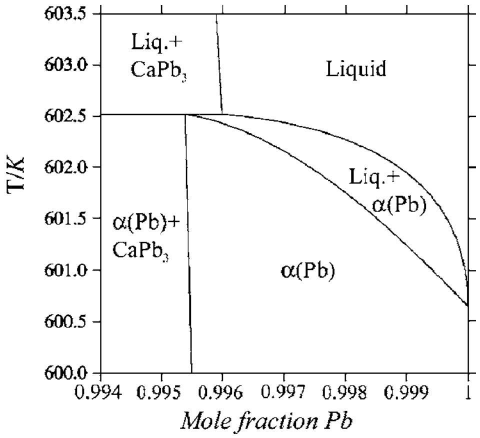

Fig. 7. Enlargement on calculated lead-rich side in Ca–Pb system.

3.4. Intermetallic compounds

The four compounds $CaPb_3$, CaPb, Ca_5Pb_3 and Ca_2Pb are considered as stoichiometric. The Gibbs energy of formation of 1 mol of formula unit is expressed as

$$G^{\mathrm{Ca}_p\mathrm{Pb}_q} - \frac{p}{p+q} H_{\mathrm{Ca}}^{\mathrm{SER}} - \frac{q}{p+q} H_{\mathrm{Pb}}^{\mathrm{SER}} = a^{\mathrm{Ca}_p\mathrm{Pb}_q} + b^{\mathrm{Ca}_p\mathrm{Pb}_q} T \qquad (13)$$

4. Results and discussion

The optimisation of the interaction parameters was carried out using the Parrot module of Thermocalc$^{\mathrm{TM}}$ software [25].

The liquid phase description was performed using the association model. This was suggested by the linear variation of the enthalpy of mixing versus composition observed by Bouirden [13] (Figs. 2 and 3). These measurements do not give any information concerning the associated complex composition. The liquid composition which generates

Table 4
Themodynamic database related to Ca–Pb system

Pure element constituents: Ca, Pb
GHSERPB
$(298.14 < T < 600.64) = -7650.085 + 101.715188T - 24.5242231T \ln T - 0.00365895T^2 - 2.439\mathrm{E}{-}07T^3$
$(600.64 < T < 1200) = -10531.115 + 154.258155T - 32.4913959T \ln T - 0.00154613T^2 + 8.05644\mathrm{E}{+}25T^{-9}$
$(1200 < T < 5000) = +4157.596 + 53.154045T - 18.9640637T \ln T - 0.00282943T^2 + 9.8144\mathrm{E}{-}08T^3 - 2696755T^{-1} + 8.05644\mathrm{E}{+}25T^{-9}$

GHSERCA
$(298.14 < T < 1115) = -4955.062 + 72.794266T - 16.3138T \ln T - 0.011110455T^2 - 133574T^{-1}$
$(1115 < T < 3000) = -107304.428 + 799.982066T - 114.292247T \ln T + 0.0237733814T^2 - 1.2438\mathrm{E}{-}06T^3 - 18245540T^{-1}$

Liquid phase constituents: Ca, Pb and Ca_2Pb (associated complex in association model [23])
${}^0G_{\mathrm{Pb}}^{\mathrm{liq}} - H_{\mathrm{Pb}}^{\mathrm{SER}} = \mathrm{GLIQPB}$
$(298.14 < T < 600.64) = +4672.157 - 7.750257T^{-6} - 6.0144\mathrm{E}{-}19T^7 + \mathrm{GHSERPB}$
$(600.64 < T < 5000) = +4853.112 - 8.066587T - 8.05644\mathrm{E}{+}25T^{-9} + \mathrm{GHSERPB}$

${}^0G_{\mathrm{Ca}}^{\mathrm{liq}} - H_{\mathrm{Ca}}^{\mathrm{SER}} = \mathrm{GLIQCA}$
$(298.14 < T < 500) = +10799.908 - 10.310466T + \mathrm{GHSERCA}$
$(500 < T < 1115) = +12793.918 - 54.496366T + 7.3263213T \ln T - 11.56082\mathrm{E}{-}03T^2 + 3.338303\mathrm{E}{-}06T^3 - 96619T^{-1} + \mathrm{GHSERCA}$
$(1115 < T < 3000) = +104649.49 - 611.059766T + 79.2922467T \ln T - 23.733814\mathrm{E}{-}03T^2 + 1.2438\mathrm{E}{-}06T^3 - 18245540T^{-1} + \mathrm{GHSERCA}$

${}^0G_{\mathrm{Ca_2Pb}}^{\mathrm{liq}} = -1.69862955\mathrm{E}{+}05 + 9.49131559T$

${}^0L_{\mathrm{Ca_2Pb,Pb}}^{\mathrm{liq}} = -65301.0497 + 8.88562411T$

${}^1L_{\mathrm{Ca_2Pb,Pb}}^{\mathrm{liq}} = -8774.044328 + 30.1452214T$

${}^0L_{\mathrm{Ca_2Pb,Ca}}^{\mathrm{liq}} = -75990.045 + 66.2967173T$

${}^1L_{\mathrm{Ca_2Pb,Ca}}^{\mathrm{liq}} = +41581.2797 - 27.7941238T$

FCC(Pb) constituents: Ca, Pb
${}^0G_{\mathrm{Pb}}^{\mathrm{FCC}} - H_{\mathrm{Pb}}^{\mathrm{SER}} = +\mathrm{GHSERPB}$

${}^0G_{\mathrm{Ca}}^{\mathrm{FCC}} - H_{\mathrm{Ca}}^{\mathrm{SER}} = +\mathrm{GHSERCA}$

${}^0L_{\mathrm{Pb,Ca}}^{\mathrm{FCC}} = -100506.392$

BCC(Ca) constituents: Ca, Pb
${}^0G_{\mathrm{Ca}}^{\mathrm{BCC}} - H_{\mathrm{Ca}}^{\mathrm{SER}} =$
$(298.14 < T < 716) = -2065.79 + 70.175889T - 11.9403T \ln T + 18.33715\mathrm{E}{-}03T^2 - 4.500217\mathrm{E}{-}06T^3 + 194152T^{-1} + \mathrm{GHSERCA}$
$(1115 < T < 3000) = +6595.537 - 70.794572T + 10.0378T \ln T - 5.08755\mathrm{E}{-}03T^2 - 389426T^{-1} + \mathrm{GHSERCA}$
$(1115 < T < 3000) = -35026.669 + 223.56698T - 29.5804512T \ln T + 8.809313\mathrm{E}{-}03T^2 - 0.460278\mathrm{E}{-}06T^3 + 7108230T^{-1} + \mathrm{GHSERCA}$

${}^0G_{\mathrm{Pb}}^{\mathrm{BCC}} - H_{\mathrm{Pb}}^{\mathrm{SER}} = 2400 - 1.1T + \mathrm{GHSERPB}$

$CaPb_3$ constituents: Ca, Pb
$G^{\mathrm{CaPb_3}} - 0.25H_{\mathrm{Ca}}^{\mathrm{SER}} - 0.75H_{\mathrm{Pb}}^{\mathrm{SER}} = -32407.5309 + 0.845325728T$

CaPb constituents: Ca, Pb
$G^{\mathrm{CaPb}} - 0.5H_{\mathrm{Ca}}^{\mathrm{SER}} - 0.5H_{\mathrm{Pb}}^{\mathrm{SER}} = -55829.3261 + 2.10392189T$

Ca_5Pb_3 constituents: Ca, Pb
$G^{\mathrm{Ca_5Pb_3}} - 0.625H_{\mathrm{Ca}}^{\mathrm{SER}} - 0.375H_{\mathrm{Pb}}^{\mathrm{SER}} = -69354.4942 + 8.93288086T$

Ca_2Pb constituents: Ca, Pb
$G^{\mathrm{Ca_2Pb}} - 0.667H_{\mathrm{Ca}}^{\mathrm{SER}} - 0.333H_{\mathrm{Pb}}^{\mathrm{SER}} = -69577.056 + 8.81587528T$

the more stable intermetallic compound (i.e. with a higher and congruent melting point) is 66.67 mol% of calcium corresponding to Ca_2Pb. Consequently, “Ca_2Pb” is the most favourable composition and was chosen as associated complex. In the present optimisation, thermodynamic data measured by Nouri et al. [17] and Fray and Kumar [18] were not used because they deviate from the experimental information obtained by Delcet et al. [16], Notin et al. [12] and Bouirden [13] which are consistent. Figs. 2–4 show that the calculated enthalpy of mixing and chemical potential are in good agreement with the experimental data.

Concerning the enthalpy of formation of the Ca–Pb intermetallic compounds measured by Bouirden [13], the accuracy obtained by this modelisation is good for the two lead-rich phases (i.e. $CaPb_3$ and CaPb). In the case of Ca_5Pb_3 and Ca_2Pb, a discrepancy between experimental and calculated values is observed (Table 3). This is due to the weak level of relevance attributed to these information in the calculation in order to keep consistency with other selected data.

The solvus curve in the lead-rich domain was determined using the data of Shumacher and Bouton [11] and Vigdorovich and Nashel’skii [10]. As shown in Figs. 5 and 6 and in Table 3, all the phase diagram data are relatively well reproduced by the present calculation.

The invariant equilibria in the lead-rich side was not clearly defined: the hypothesis of a peritectic invariant has been accepted in accordance with Shumacher and Bouton [11] and Notin et al. [12]. Consequently, the invariant temperature introduced in the optimisation was $T_{\text{inv}} = 601.9\,\text{K}$, i.e. about 1 K higher than the pure lead melting point. Fig. 7 shows the peritectic situation obtained by the present calculation (Table 3).

Finally, the parameter set adjusted from all the selected experimental information is reported in Table 4.

5. Conclusions and outlooks

The modelling of the Ca–Pb system has led to a good reproduction of the main thermodynamic and phase diagram data collected from the literature. The good results obtained during this optimisation are essentially due to the consideration of the existence of an associated complex in the liquid phase. This was later described using the association model which allows to calculate the parameters of the liquid phase.

Considering the similarities between the Ca–Pb and Ca–Sn systems, the latter will be studied in the same way in order to obtain the thermodynamic description of the ternary system Ca–Pb–Sn. Its description will be interesting in order to explain and optimise the hardening process occurring in these alloys used in the acid batteries.

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