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CALORIMETRIC INVESTIGATION OF THE Cu–Sn–Bi LEAD-FREE SOLDER SYSTEM

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The thermodynamic properties play a crucial role in the development of new solder materials. In this work a calorimetric investigation of the ternary Cu–Sn–Bi system was carried out by using a Calvet-type calorimeter in order to obtain the molar limiting partial enthalpy of Cu in liquid Sn–Bi alloys with different compositions. The molar limiting partial enthalpy of Cu at 820 K was determined in the Sn–75 at% Bi, Sn–43 at% Bi and Sn–26 at% Bi liquid bath showing an endothermic behaviour. The results are compared with the literature data available for Cu in the pure liquid Bi and Sn and then discussed.

Keywords: *dissolution calorimetry, enthalpy, lead-free solder, liquid alloys*

Introduction

In recent years the electronic industry has been involved in the development of a new generation of solder interconnects in order to replace the Pb–Sn with Pb-free solder alloys. This is due to legal, environmental and technological factors which call for alternative lead-free soldering materials. For these reasons a large number of Pb-free solder alloys have been proposed [1]. Among the huge number of proposed systems, many of the most promising lead-free alloys are tin-based systems [2, 3]. The alloying elements should be chosen in order to maintain the low melting point and ensure mechanical, wetting, and transport properties, similar to the Pb–Sn alloy. From this point of view the Sn–Bi system is one of the most interesting ones. It has been developed for step-by-step soldering processes and it is suitable for soldering temperature-sensitive components and substrates. However, this system presents some drawbacks concerning firstly the mechanical properties e.g. potential embrittlement and poor thermal fatigue properties. Moreover, when compared to the Pb–Sn solder, Sn–Bi solder shows a higher strength and superior creep resistance. Furthermore the addition of Bi to Sn-based alloy can improve wetting between solders and substrates. Thus, the Sn–Bi alloys are a promising lead-free solder system, although the availability of Bi could be limited by the restriction on Pb, because the primary source of Bi is a by-product in Pb refining [4]. Some reports anyway estimate that the Bi supply will be enough for solders even with Sn–43 at% Bi eutectic solder system.

The solder process consists in the formation of joints between the molten solder, the interconnects and a solid metal surface. Therefore, the choice of the substrate is fundamental in the study of reliability of the solder joints. The most common substrates used in the electronic devices are Cu, Ni, Au and Pd. The formation of a solder joint is a complex problem which involves many different chemical and physical processes as, for example, the melting process, the spreading of the liquid alloy over the substrate, the diffusion between the liquid and solid, or the formation of possible intermetallic compounds at the interface which can characterise the behaviour of the metal–metal system [5].

For a comprehensive understanding of all these processes, information on the phase equilibria in the intermetallic systems generated by the soldering process are indispensable, and should be supported by the relevant information on thermodynamic properties.

Even if a huge number of lead free solder candidates are ternary or higher order alloys and, together with one or more contact materials these result in multicomponent systems, the experimental characterization of such complex systems is very difficult and time consuming. However, the well-known CALPHAD procedure [6] can provide a way to calculate phase equilibria in complex systems based on reliable experimental data. For this reason the knowledge of the thermodynamic data of basic

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binary and ternary systems is crucial in the development of new solder materials.

In this work we investigated the thermodynamic properties of the Cu–Sn–Bi ternary system by using a Calvet-type calorimeter. We measured the molar partial enthalpy of solid Cu in liquid Sn–Bi at different compositions and we evaluated the molar limiting partial enthalpy. These data may be the first step in the characterization of more complex systems of a great technological interest.

Literature data

Thermodynamic data on the Cu–Sn–Bi and binary subsystems are available in literature and are here reported in order to have a more comprehensive understanding of the characterization of this system.

Cu–Sn

The Cu–Sn system is a well known intermetallic system (brass alloys) and a great number of experimental investigations have been performed on it. The existing phase diagram is mainly based on the work by Raynor [7]. Thermochemical data and assessment of the phase diagram were published by Saunders and Miodownik [8] and Shim *et al.* [9]. Several calorimetric investigations of the molar limiting partial enthalpy of mixing of Cu in liquid Sn have been published. The measurements were carried out in a wide temperature range between 500 and 1100 K. Deneuville *et al.* [10] and Yassin and Castanet [11] have both published compilations of the experimental data of the limiting partial enthalpy of Cu in liquid Sn. These papers show a temperature dependence of the enthalpy of mixing in this system. Recently, Ipser *et al.* [12] and Flandorfer *et al.* [13], have fully investigated the thermodynamic properties of lead-free solder materials. They measured the integral enthalpies of mixing of the Cu–Sn system at different temperatures and also reported the partial enthalpy of Cu in liquid Sn in a temperature range between 773 and 1073 K confirming the temperature dependence found in the earlier works. The authors [12, 13] also report the formation enthalpy of some Cu–Sn binary intermetallic phases: Cu_3Sn (ε-phase), $Cu_{41}Sn_{11}$ (δ-phase) and Cu_6Sn_5 (η-phase). They obtained slightly exothermic values ranging from -5.7 ± 1 kJ mol^{-1} for δ-phase to -8.2 ± 1 kJ mol^{-1} for ε-phase. These results show that the ε-phase is the most energetically favoured.

Cu–Bi

The binary Cu–Bi system has also been widely investigated by calorimetry and emf measurements, since 1930 [14]. Chakrabarti and Laughlin [15] then collected the thermodynamic studies reported up to 1982. The resulting enthalpies of mixing of liquid alloys are generally endothermic but there is a discrepancy between the different experimental results. Niemelä *et al.* [16] and Teppo *et al.* [17] published two different thermodynamic assessments. The enthalpies of mixing does not show a significant temperature dependence.

Sn–Bi

Yazawa *et al.* [18] and Sharkey and Pool [19], investigated the thermodynamic properties of this system by calorimetric measurements, while emf measurements were carried out by Seltz and Dunkerley [20]. Hultgren *et al.* [21] reported a review of all the thermodynamic values available. The integral thermodynamic quantities of the liquid system were assessed by Cho and Ochoa [22]. More recently Asryan and Mikula [23] and Cho *et al.* [24], have investigated this system by emf method using liquid and solid electrolytes respectively. Asryan and Mikula [23] showed that the integral mixing enthalpy *vs.* composition is asymmetric. The phase diagram has been reviewed and thermodynamically calculated by Lee *et al.* [25].

Experimental

The molar partial enthalpy of Cu ($\Delta\overline{H}_{\mathrm{m,Cu}}$) in different liquid Sn–Bi alloys was determined at different temperatures. The $\Delta\overline{H}_{\mathrm{m,Cu}}$ at 820 K was measured in Sn–75 at% Bi, Sn–43 at% Bi and Sn–26 at% Bi compositions, while a liquid Sn–43 at% Bi bath was used at 920 K.

The Cu content ranged from 0 to 1 at% in order to maintain infinite dilution conditions necessary for the determination of the molar limiting partial enthalpy.

The sample were prepared from high purity metals (99.999%) mechanically scratched and chemically cleaned. In particular the Cu was cleaned in a H_2-flow at 423 K for 2 h to remove the oxide layer. Small pieces of Cu (mass between 80 and 300 mg) were added into a liquid bath formed by Sn–Bi (with mass around 20 g).

The experiments of this work were performed using a high-temperature Calvet calorimeter generously supplied by the ‘Thermodynamiques des Systèmes Métalliques’ laboratory, and settled in our laboratory by Jean Pierre Bros, in the framework of the agreement between the Aix-Marseille I University and Genoa University. This calorimeter is a home-made instrument ad hoc designed for thermodynamic investigations of metallic systems. Two calorimetric cells are located inside the alumina block surrounded by two large cylindrical alumina shields in order to guarantee the isothermal conditions, and heated by four Kanthal resistors. The furnace is controlled by an automatic device in order to maintain constant the temperature of the apparatus in a range of ±0.5 K; the furnace can reach a temperature up to 1300 K. The two cells, surrounded by thermopiles, are maintained in symmetrical conditions (Ar flux, crucible, etc.), although one cell is used as the reference cell.

Each thermopile consists of a cylindrical thin-walled alumina tube which is closed at the bottom and surrounded by 320 Pt–Pt 10% Rh thermocouples. The output of the thermopiles is proportional to the amount of heat flowing from the cell to the block. The two thermopiles are linked in differential mode in order to acquire the difference in the thermal flows. The signal difference of the two thermopiles is recorded in real time (scan rate 1 s^{-1}) as a function of time using an appropriate home-made software (CALMAA) developed in the LabView® environment together with the room and calorimetric temperatures.

A quartz tube containing the sample crucible is placed inside the measuring cell. It is connected to a gas line which makes it possible to evacuate and flush the sample with Ar. Our measurements were carried out under a pure argon (99.999%) flow of 6 L h^{-1}. The crucible is made of graphite (Carbone Lorraine high purity grade) in order to avoid any reaction of the metals with the container and also to have a reduction surrounding atmosphere. A Ta foil getter is placed under the crucible in order to reduce the oxygen partial pressure.

As mentioned above, small pieces of solid metal were dropped into the liquid metallic bath from room temperature to the calorimeter temperature. After each series of drops the calorimeter was calibrated by adding 5 pieces of α-Al_2O_3 standard from NIST (National Institute of Standard and Technology, Gaithersburg, MD).

The signals were integrated by appropriate software developed by using the analysing software Igor Pro®. The numerical integration was carried out by the rectangular method between the signal acquired and the baseline.

During each drop, inside the measuring cell the following reaction takes place

$$n_i\mathrm{A}(p^0,T_r,s) + m\mathrm{BC}(p^0,T_c,l) \rightarrow \mathrm{A_xB_yC_z}(p^0,T_c,l) \qquad (1)$$

where x, y, z are molar fractions, n_i is the number of moles of the element A, solid and at room temperature T_r, added at each drop, m is the number of moles of the liquid BC alloys contained in the crucible at the calorimeter temperature T_c.

During the reaction (1) a heat Q_p is exchanged in the measuring cell

$$Q_P = n_i\Delta H_1 + n_i\Delta H_2 + (n_i + m)\Delta_{mix}H_m \qquad (2)$$

All the enthalpies are considered in the standard state, but the apex ° is omitted, for clarity, in the text. $\Delta_{mix}H_m$ is the integral enthalpy of mixing, and ΔH_1 and ΔH_2 are given by

$$\Delta H_1 = \int_{T_r}^{T_m} c_{P,m}(A,s)\mathrm{d}T$$
$$\Delta H_2 = \left[\int_{T_m}^{T_c} c_{P,m}(A,l)\mathrm{d}T + \Delta_{fus}H(A)\right] \qquad (3)$$

corresponding respectively to the heating enthalpy of A from room to melting temperature, and to the sum of the heating enthalpy of A from the melting temperature to the calorimeter temperature with the addition of the melting enthalpy of A. These two contributions were calculated using the Dinsdale's polynomials [26].

On the other hand, the molar partial enthalpy of the C element added to the binary metallic bath can be determined by

$$\Delta\overline{H}_{m,C} = \frac{Q_P}{n_i} - n_i(\Delta H_1 + \Delta H_2) \qquad (4)$$

After each experiment the samples were took out and cooled to room temperature and the microstructure and the composition of the samples were checked by scanning electron microscope (SEM) equipped with electron probe microanalysis (EPMA).

It may be worthwhile to underline that the Calvet-type calorimeter has often been employed, as in our experiments, to obtain partial and integral enthalpies; furthermore this calorimeter and method can be used to determine the phase boundaries of the phase diagram of the system under investigation [27–29].

Results and discussion

The results of the molar partial enthalpy of Cu at T=820 K in three different liquid Sn–Bi alloy compositions are reported in Table 1 together with the results obtained at 920 K for the Sn–43 at% Bi liquid alloy. Table 1 reports the mass of solid Cu added at each drop, the resulting Cu amount in at% and the

Table 1 Molar partial enthalpies of Cu ($\Delta\overline{H}_{m,Cu}$) in different liquid Bi–Sn bath and temperatures with the corresponding mass of Cu and the resulting at% of Cu. For each run the masses of Bi (m_{Bi}) and Sn (m_{Sn}) present in the liquid bath, and the total mass (m_{tot}) of the liquid alloy are reported

m_{Cu}/g	Cu/at%	$\Delta\overline{H}_{m,Cu}$/kJ mol^{-1} (±0.5)
Sn–75 at% Bi, T=820 K		
m_{Bi}=16.8171 g; m_{Sn}=3.1858 g; m_{tot}=20.0029 g		
0.0083	0.10	13.3
0.0110	0.25	11.7
0.0122	0.40	12.1
0.0144	0.57	11.9
0.0155	0.76	11.7
m_{Bi}=16.8168 g; m_{Sn}=3.1836 g; m_{tot}=20.0004 g		
0.0094	0.12	13.4
0.0108	0.24	15.4
0.0120	0.39	12.8
0.0127	0.52	12.8
m_{Bi}=16.8130 g; m_{Sn}=3.1816 g; m_{tot}=19.9946 g		
0.0082	0.10	14.8
0.0109	0.25	13.5
0.0111	0.37	13.2
0.0130	0.53	12.8
Sn–43 at% Bi, T=820 K		
m_{Bi}=11.4087 g; m_{Sn}=8.5902 g; m_{tot}=19.9989 g		
0.0251	0.61	6.6
0.0302	0.98	8.7
m_{Bi}=11.4088 g; m_{Sn}=8.5094 g; m_{tot}=19.9182 g		
0.0237	0.29	7.5
0.0280	0.64	7.3
0.0299	1.01	5.8
m_{Bi}=11.4088 g; m_{Sn}=8.5901 g; m_{tot}=19.9989 g		
0.0198	0.24	8.7
0.0283	0.59	6.1
0.0287	0.94	6.0
m_{Bi}=11.4107 g; m_{Sn}=8.5918 g; m_{tot}=20.0025 g		
0.0109	0.13	11.0
0.0163	0.34	8.6
0.0215	0.60	7.2
0.0281	0.94	6.0
Sn–26 at% Bi, T=820 K		
m_{Bi}=7.7226 g; m_{Sn}=12.2792 g; m_{tot}=20.0018 g		
0.0095	0.12	4.2
0.0124	0.27	3.9
0.0143	0.45	3.9
0.0191	0.68	3.6
m_{Bi}=7.7196 g; m_{Sn}=12.2805 g; m_{tot}=20.0001 g		
0.0090	0.11	2.6
0.0101	0.24	5.6
0.0123	0.39	5.5
0.0144	0.56	4.4
m_{Bi}=7.7207 g; m_{Sn}=12.2809 g; m_{tot}=20.0016 g		
0.0065	0.08	5.3
0.0070	0.17	5.8
0.0112	0.31	5.5
0.0139	0.48	4.3
0.0184	0.70	4.2
m_{Bi}=7.7149 g; m_{Sn}=12.2786 g; m_{tot}=19.9935 g		
0.0052	0.06	3.1
0.0075	0.16	3.7
0.0110	0.29	5.3
0.0131	0.45	5.6
0.0137	0.62	4.0
Sn–43 at% Bi, T=920 K		
m_{Bi}=11.4089 g; m_{Sn}=8.5912 g; m_{tot}=20.0001 g		
0.0129	0.16	13.3
0.0157	0.35	10.1
0.0171	0.56	7.1
0.0243	0.89	9.4
0.0268	1.19	7.9
m_{Bi}=11.4102 g; m_{Sn}=8.5900 g; m_{tot}=20.0002 g		
0.0096	0.12	11.4
0.0123	0.27	9.9
0.0130	0.43	9.5
0.0148	0.61	9.3
0.0242	0.91	7.9
m_{Bi}=11.4095 g; m_{Sn}=8.5900 g; m_{tot}=19.9995 g		
0.0084	0.10	11.8
0.0120	0.25	10.9
0.0156	0.44	9.5
0.0165	0.65	7.0

Table 1 Continued

m_{Cu}/g	Cu/at%	$\Delta\overline{H}_{m,Cu}$/kJ mol^{-1} (±0.5)
Sn–43 at% Bi, T=920 K		
m_{Bi}=11.4092 g; m_{Sn}=8.5995 g; m_{tot}=20.0087 g		
0.0095	0.12	10.7
0.0120	0.27	8.1
0.0136	0.43	10.5
0.0148	0.61	9.5
0.0178	0.83	7.5
m_{Bi}=11.4070 g; m_{Sn}=8.5903 g; m_{tot}=19.9973 g		
0.0110	0.14	9.1
0.0115	0.28	9.1
0.0126	0.43	8.1
0.0196	0.67	9.1
m_{Bi}=11.4094 g; m_{Sn}=8.5912 g; m_{tot}=20.0006 g		
0.0102	0.13	9.6
0.0107	0.26	10.0
0.0132	0.42	12.2
0.0139	0.59	10.7
0.0146	0.77	9.4

value of the molar partial enthalpy. The measurements were carried out in different runs in order to have the necessary number of values to obtain a suitable regression function giving at x_{Cu}=0, the values of the molar limiting partial enthalpy ($\Delta\overline{H}^{\infty}_{m,Cu}$). Figure 1 reports the results of the molar partial enthalpy of Cu in the liquid Sn–Bi alloys at three different compositions, and T=820 K together with the corresponding linear fit.

The resulting values of the molar limiting partial enthalpy obtained are: +14.2±0.5, +8.6±0.5 and +5.6±0.5 kJ mol^{-1} for Sn–75 at% Bi, Sn–43 at% Bi and Sn–26 at% Bi respectively. These endothermic values are in agreement with the literature data of the integral enthalpy of mixing of Cu–Sn–Bi reported by Ipser *et al.* [12]. The authors report the isoenthalpy plot of the Cu–Sn–Bi ternary system from which it is possible to deduce a parabolic convex trend of the integral enthalpy of mixing along the isopleth sections considered. From this plot it is possible to obtain a graphically qualitative evaluation of the molar limiting partial enthalpy which is endothermic as in our experimental work.

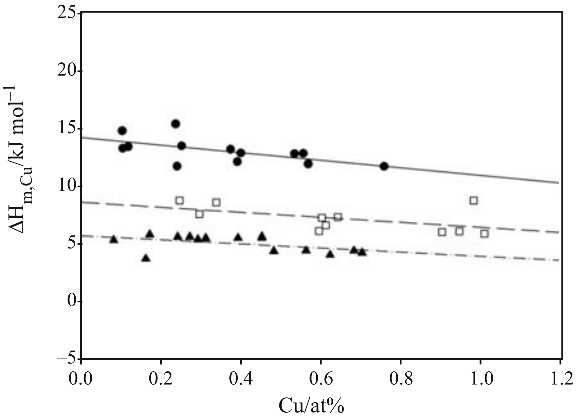

Fig. 1 Molar partial enthalpy of Cu at T=820 K as a function of at% Cu for three different compositions of the liquid Sn–Bi bath: ● – Sn–75 at% Bi, □ – Sn–43 at% Bi, ▲– Sn–26 at% Bi

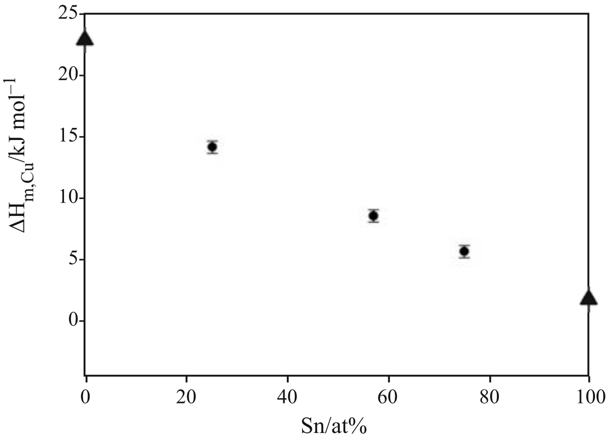

Fig. 2 Molar limiting partial enthalpy of Cu at T=820 K as a function of Sn amount in the Sn–Bi liquid bath alloys. Experimental data: ● – this work, ▲ – literature data for Cu in pure Bi [21] and Cu in pure Sn [12]

Figure 2 shows the values of the molar limiting partial enthalpy of Cu as a function of the Sn content in the Sn–Bi liquid alloys together with the literature data for Cu in pure molten Bi [21] and Sn [12, 13]. Our results are in good agreement with the values available for pure metals and show that the molar limiting partial enthalpy decreases with the increasing of the Sn content in the liquid alloy.

After the calorimetric measurements, the thermodynamic equilibrium of the samples was checked by SEM-EPMA analysis. The characterization of the microstructure of these samples confirmed that the complete dissolution of Cu in the Sn–Bi liquid bath took place. All samples showed the presence of the Sn–Bi eutectic mixture together with the formation of very small quantities of the η-Cu_6Sn_5 and ε-Cu_3Sn phases formed during the cooling of the liquid bath.

The molar limiting partial enthalpy of Cu in Sn–43 at% Bi was also measured at T=920 K in order to investigate a possible temperature dependence. The resulting value +10.6±0.5 kJ mol^{-1} shows a very slight increase in the molar limiting partial enthalpy with the increasing of the temperature. Ipser *et al.* [12] reported a similar behaviour for Cu in pure Sn and it is known from the literature that the enthalpy of mixing of Cu–Bi does not show any temperature dependence.

Conclusions

The molar partial enthalpy of Cu at T=820 K was measured by calorimetry in Sn–75 at% Bi, Sn–43 at% Bi and Sn–26 at% Bi liquid alloys and at T=920 K in Sn–43 at% Bi. By linear regression the values of the molar limiting partial enthalpy of Cu ($\Delta\overline{H}^{\infty}_{\mathrm{m,Cu}}$) were obtained resulting slightly positive and ranging from +5.6±0.5 to +14.2±0.5 kJ mol^{-1}. Our experimental results are in good agreement with the literature data available for Cu in pure liquid Bi and in pure liquid Sn, and they show a monotonic decrease from +23 kJ mol^{-1} for Cu in pure liquid Bi to +0.9 kJ mol^{-1} for Cu in pure liquid Sn when the Sn content increases in the liquid Bi–Sn alloys. A slight temperature dependence of the molar limiting partial enthalpy was also observed for the dissolution of Cu in the Sn–43 at% Bi liquid alloy.

These results suggest that the presence of Bi can reduce the dissolution of Cu in the alloy, while an increase of the Sn amount promotes the Cu dissolution. For this reason, the Bi could be used in the Sn-based solder in order to modulate the dissolution of the solid Cu substrate inside the liquid alloy during the spreading process [30].

The data obtained in the present investigation could be useful for a further thermodynamic optimization and assessment of the Cu–Sn–Bi system, according to the CALPHAD method.

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References

1 M. Abtew and G. Selvaduray, Mater. Sci. Eng., 27 (2000) 95.
2 P. T. Vianco and D. R. Frear, JOM, 45 (1993) 14.
3 D. Zivkovic, A. Milosavljevic, A. Mitovski and B. Marjanovic, J. Therm. Anal. Cal., 89 (2007) 137.
4 K. Zeng and K. N. Tu, Mater. Sci. Eng. R., 38 (2002) 55.
5 J. A. Warren, W. J. Boettinger and A. R. Roosen, Acta Mater., 46 (1998) 3247.
6 N. Saunders and A. P. Miodownik, CALPHAD Calculation of Phase Diagrams, Pergamon Materials Series, Cambridge 1998.
7 G.-V. Raynor, Annotated Equilibrium Diagram Series, No. 2, The Institute of Metals, London 1944.
8 N. Saunders and A. P. Miodownik, Bull. Alloy Phase Diagrams, 11 (1990) 278.
9 J.-H. Shim and C.-S. Oh, Z. Metallkd., 87 (1996) 205.
10 J.-L. Deneuville, C. Chatillon-Colinet and J.-C. Mathieu, Ann. Chem. Sci. Mater., 4 (1979) 431.
11 A. Yassin and R. Castanet, J. Alloys Compd., 307 (2000) 191.
12 H. Ipser, H. Flandorfer, C. Luef, C. Schmetterer and U. Saeed, J. Mater. Sci: Mater. Electron., 18 (2007) 3.
13 H. Flandorfer, U. Saeed, C. Luef, A. Sabbar and H. Ipser, Thermochim. Acta, 459 (2007) 34.
14 M. Kawakami, Sci. Repts. Tohoku Imp. Univ., 19 (1930) 521.
15 D. Chakrabarti and D. Laughlin, Bull. Alloy Phase Diagrams, 5 (1984) 148.
16 J. Niemelä, G. Effenberg, K. Hack and P. Spencer, CALPHAD, 10 (1986) 77.
17 O. Teppo, J. Niemelä and P. Taskinen, Thermochim. Acta, 173 (1990) 137.
18 A. Yazawa, T. Kawashima and K. Itagaki, J. Jpn. Inst. Met., 32 (1968) 1281.
19 R. L. Sharkey and M. J. Pool, Metall. Trans., 3 (1972) 1773.
20 H. Seltz and F. J. Dunkerley, J. Am. Chem. Soc., 64 (1942) 1392.
21 R. Hultgren, P.S. Desai, D. T. Hawkins, M. Gleiser and K. K. Kelley, Selected Values of the Thermodynamic Properties of Binary Alloys, ASM, Metals Park, OH 1973.
22 S.-A. Cho and J. L. Ochoa, Metall. Mater. Trans. A, 29 (1998) 1965.
23 N. Asryan and A. Mikula, Z. Metallkd., 95 (2004) 132.
24 S.-A. Cho, R. Camisotti and M. H. Vélez, Metall. Trans. B, 21B (1990) 87.
25 B. J. Lee, C. S. Oh and J. H. Shim, J. Electr. Mater., 25 (1996) 983.
26 A. T. Dinsdale, CALPHAD, 13 (1991) 317.
27 R. Castanet, J. Phase Equilib., 15 (1994) 339.
28 M. H. Hamedi and J.-P. E. Grolier, J. Therm. Anal. Cal., 89 (2007) 87.
29 W. Zielenkiewicz, J. Therm. Anal. Cal., 91 (2008) 663.
30 F. Gnecco, E. Ricci, S. Amore, D. Giuranno, G. Borzone, G. Zanicchi and R. Novakovic, Int. J. Adhes. Adhes., 27 (2007) 409.

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