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# Integral enthalpy of mixing of the liquid ternary Au–Cu–Sn system

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## Abstract

The integral enthalpy of mixing of the ternary Au–Cu–Sn has been determined with a Calvet type calorimeter at 6 different cross sections at 1273 K. The substitutional solution model of Redlich–Kister–Muggianu was used for a least square fit of the experimental data in order to get an analytical expression for the integral enthalpy of mixing. The ternary extrapolation models of Kohler, Muggianu and Toop were used to calculate the integral enthalpy of mixing and to compare measured and extrapolated values. Additional calculations of the integral enthalpy of mixing using the Chou model have been performed. With the calculated data, the iso-enthalpy lines have been determined using the Redlich–Kister–Muggianu model. A comparison of the data has been made.

*Keywords:* Thermodynamic; Calorimetry; Enthalpy of mixing

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## 1. Introduction

For all metallurgical processes the knowledge of the thermodynamic properties of alloy systems is necessary. The possibility of a reaction, the stability of phases, the reaction path, and many more physical properties depend on it. It can also be used for the calculation of the phase diagram and various alloy properties like the wetting behaviour and the surface tension.

The aim of thermodynamic investigations is the determination of the partial and integral quantities of metallic systems depending on concentration, temperature and pressure. Various methods are available and can be chosen, depending on the system and on the properties that shall be determined.

With this knowledge, the properties of higher order systems can be calculated. Various extrapolation and calculation models are available and are widely used [1–4].

One quantity that has to be known is the integral enthalpy of mixing, which can be determined by calorimetric measurements.

## 2. Experimental procedure

For the determination of the enthalpy of mixing of the ternary system a Calvet type microcalorimeter (Fa. SETARAM; Lyon) was used. The surrounding of the furnace has been adapted to our requirements [5]. The measurements were performed at 1273 K by a direct drop method with an automatic sample drop device under Ar flow.

The calorimeter furnace is a twin calorimeter with two separate calorimeter cells, surrounded by thermopiles (TPs) with more than 200 pairs of thermocouples each. Each TP is mounted as a ring formed by two concentric cylinders measuring the temperature difference between the inner and outer surface. In this way, the EMF of a TP gives direct information on the heat flow absorbed or produced by the calorimeter cell.

The calorimeter cells are located in the large calorimeter block made of alumina and kanthal which is surrounded by thermal isolating bricks and the electric resistance heating. The cells are connected in opposition, and variations of the temperature differences down to $10^{-5}$ K can be detected. The oscillations of temperature of the furnace will be compensated. One cell is usually permanently loaded with a block of about 25 g alumina and serves as the reference cell. The alumina block provides similar heat capacity and thermal conductivity of the two cells in order to avoid secondary effects on the calorimeter signal. The furnace further contains two single thermocouples. One measures the sample temperature and the other is used to control the furnace temperature together with a Eurotherm 818 Controller.

All thermocouples are of type S (Pt/Pt-10 wt.% Rh). The furnace can be operated from ambient temperature up to 1100 °C. The measuring cell is charged with a quartz tube which contains

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the sample crucible. This quartz tube is connected to the automatic sample introducing unit. The entire system is gas tight and can be evacuated and flushed with a protective gas. Our measurements were performed under a continuous argon flow (30 cm$^3$/min). The gained signals were recorded and automatically integrated.

The calorimeter is described in more detail in [5]. After each series of drops the calorimeter was calibrated by adding 5 pieces of α-$Al_2O_3$ standard of NIST (National Institute of Standards and Technology, Gaithersburg, MD). The temperature in the drop device (DT) and the furnace temperature (FT) were measured before each drop.

The measured enthalpy is given by

$$H_{\text{Signal}} = n_i \cdot (H_{\text{Sample,FT}} - H_{\text{Sample,DT}}) + \Delta H_{\text{Reaction}} \qquad (1)$$

$n_i$ is the amount of the added sample. The relative enthalpy $H_{\text{Sample, FT}} - H_{\text{Sample, DT}}$ was calculated using the polynomials from Dinsdale [6] for the thermodynamic data of pure elements. The approximate partial enthalpies of Au and Cu in the ternary Au–Cu–Sn system could be determined directly by

$$\overline{\Delta H_i} \approx \left(\frac{H_{\text{Signal}}}{n_i}\right) - \left(H_{\text{Sample,FT}} - H_{\text{Sample,DT}}\right) \qquad (2)$$

The integral enthalpy of mixing was calculated by

$$\Delta H_{\text{Mix}} = \Delta H_{\text{bin}} + \sum\left(\overline{\Delta H_i} \times \frac{1}{(n + \sum n_i)}\right) \qquad (3)$$

$n$ is the molar amount and $\Delta H_{\text{bin}}$ the integral enthalpy of the metals propounded in the crucible. All metals were of high purity (99.98+ %). Sn rod (Johnson Matthey GmbH, Karlsruhe, Germany) was cleaned with a fine sand paper before using. Copper (Fa. GoodFellow, Cambridge, England) wire was heated for 2 h at 200 °C under hydrogen flow. Au was a product of Ögussa, Vienna, Austria. For the measurement of the ternary system as a first step, gold was added to the molten binary Cu–Sn alloys in the composition range from 0 to 50 at.% Au. In a second step, copper was dropped into the molten Au–Sn system in the composition range from 0 to 50 at.% Cu. The compositions, which were measured at 1273 K are shown in Fig. 1. The time interval between the drops was 40 min.

## 3. Binary systems

### 3.1. Gold–copper system

A compilation of this binary system has been given by Hultgren et al. [7]. More recent investigations have been performed by Topor and Kleppa [8] and Fitzner et al. [9].

### 3.2. Gold–tin system

For the integration the binary data given by Hultgren et al. [7] at 823 K were used. A more recent investigation of the binary systems has been done by Hayer et al. [10]. The difference between the investigated data was minimal. A summary concerning the binary Au–Sn system has been published by Okamoto and Massalski [11].

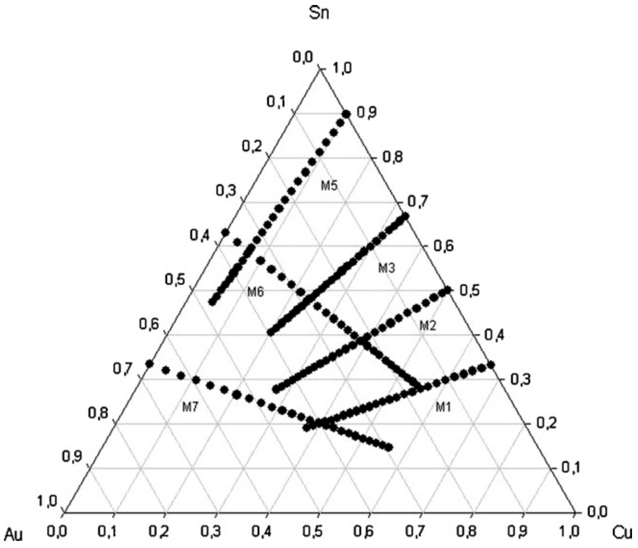

Fig. 1. Measured cross sections of the ternary Au–Cu–Sn system (M4–M3).

### 3.3. Copper–tin system

Hultgren et al. [7] evaluated the thermodynamic properties of this system. A critical analysis of the binary phase diagram has been done by Miodownik [12].

The partial free energy of formation of α-Cu solid solution in the Cu–Sn system and the intermetallic phases were detected using solid-state galvanic cells and bonding contributions to the partial free energy of formation were calculated by Predel and Schallner [13].

Predel and Schallner [14] investigated furthermore the thermodynamic properties with an EMF method. A thermodynamic assessment has been done by Shim et al. [15] using the CALPHAD method. Recently calorimetrical measurements were performed by Luef et al. [16]. His values were used as starting values for the integration.

## 4. Extrapolation models

Three different extrapolation models were used to calculate the enthalpy of mixing in the ternary systems. These values were compared with the experimental values. The models from Kohler [1] and Muggianu et al. [2] use simple symmetric extrapolation models to predict ternary data based on binary data, whereas the Toop model [3] is an asymmetric one.

## 5. Ternary modelling

The experimental data were treated with a least squares fit using the Redlich–Kister–Muggianu polynomial (Eq. (4)) given by Ansara and Dupin [17] for substitutional solutions, which was also used for the calculation of the iso-enthalpy curves of the integral molar enthalpy of mixing, of the ternary Au–Cu–Sn system.

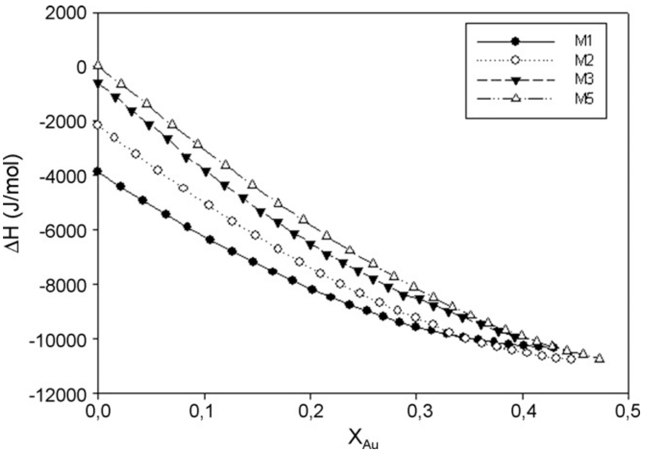

Fig. 2. Integral enthalpy of mixing of the ternary Au–Cu–Sn system at 1273 K at the cross section Cu:Sn, with Au added (M4–M3).

$i$ and $j$ are equal 1, 2 and 3 representing the elements A, B and C, in the case of this work Au, Cu and Sn. $L_{i:j}^{(n)}$ ($\nu = 0, 1, \ldots$) are the interaction parameters of the binary Au–Cu, Au–Sn and Cu–Sn systems taken from Refs. [7,8,16].

$$\Delta H_{\text{Mix}} = \sum_i \sum_{j\rangle i} [xixj \sum_{\nu} L(\nu)i : j(x_i - x_j)^{\nu}] + x_A x_B x_C (M^{(0)}_{A:B:C} x_A + M^{(1)}_{A:B:C} x_B + M^{(2)}_{A:B:C} x_C) \qquad (4)$$

The last term represents the additional enthalpy of mixing due to the ternary interactions where $M^{(a)}_{A:B:C}$ are the excess ternary interaction parameters.

## 6. Calculation

For the calculation of the integral enthalpy of mixing the model given by Chou et al. [4] has been used. The binary interaction parameter as shown in Table 3, has been used for the calculation of the $\Delta H$ values of the ternary system. The model of Chou et al. [4] is different from the extrapolation models, because a ternary interaction parameter is used and therefore

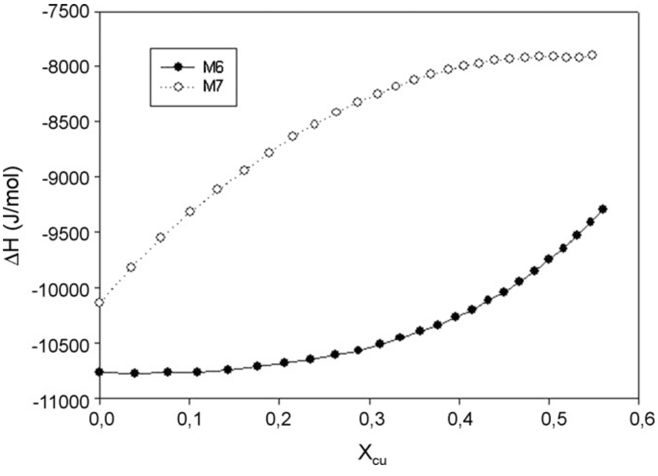

Fig. 3. Integral enthalpy of mixing of the ternary Au–Cu–Sn system at 1273 K at the cross section Au:Sn, with Cu added.

Table 1
Partial and integral enthalpies of mixing resulting from calorimetric measurements at different constant ratios of Cu:Sn with Au dropped from 0 to 55 at.% in the ternary system Au–Cu–Sn at 1273 K

| $m_{Au}$ (mg) | $x_{Au}$ | $\overline{\Delta H}_{Au}$ (J/mol) | $\Delta H_M$ (J/mol) |
|---|---|---|---|
| M1 | | | |
| $m_{Cu}$ = 261.91 mg, $m_{Sn}$ = 243.75 mg | | | |
| | | | −3874 |
| 26.14 | 0.0210 | −29 131 | −4405 |
| 28.29 | 0.0428 | −28 211 | −4935 |
| 28.50 | 0.0638 | −27 577 | −5432 |
| 29.34 | 0.0845 | −26 597 | −5899 |
| 31.50 | 0.1057 | −26 170 | −6369 |
| 32.04 | 0.1263 | −24 823 | −6793 |
| 32.21 | 0.1460 | −24 123 | −7185 |
| 32.25 | 0.1649 | −23 067 | −7537 |
| 33.29 | 0.1836 | −22 791 | −7878 |
| 34.25 | 0.2019 | −21 809 | −8191 |
| 34.62 | 0.2197 | −20 880 | −8473 |
| 35.08 | 0.2368 | −20 932 | −8747 |
| 35.17 | 0.2533 | −19 358 | −8976 |
| 35.61 | 0.2693 | −19 319 | −9197 |
| 35.83 | 0.2847 | −17 953 | −9382 |
| 36.55 | 0.2997 | −17 368 | −9550 |
| 37.38 | 0.3145 | −16 538 | −9697 |
| 37.73 | 0.3288 | −15 870 | −9825 |
| 42.78 | 0.3442 | −15 199 | −9949 |
| 43.29 | 0.3592 | −14 255 | −10 048 |
| 43.44 | 0.3735 | −13 844 | −10 132 |
| 44.78 | 0.3877 | −13 045 | −10 198 |
| 45.78 | 0.4015 | −12 583 | −10 252 |
| 50.43 | 0.4159 | −11 781 | −10 289 |
| 51.99 | 0.4302 | −11 382 | −10 315 |
| M2 | | | |
| $m_{Cu}$ = 205.41 mg, $m_{Sn}$ = 384.50 mg | | | |
| | | | −1975 |
| 20.17 | 0.0156 | −32 255 | −2447 |
| 27.14 | 0.0358 | −31 572 | −3044 |
| 28.57 | 0.0562 | −30 900 | −3634 |
| 35.83 | 0.0806 | −29 738 | −4308 |
| 36.74 | 0.1043 | −28 937 | −4944 |
| 36.76 | 0.1269 | −28 007 | −5525 |
| 37.54 | 0.1487 | −26 433 | −6049 |
| 38.27 | 0.1700 | −26 464 | −6557 |
| 38.51 | 0.1903 | −26 225 | −7039 |
| 39.15 | 0.2099 | −24 397 | −7460 |
| 39.16 | 0.2286 | −23 428 | −7838 |
| 41.09 | 0.2473 | −23 060 | −8207 |
| 41.54 | 0.2654 | −22 030 | −8538 |
| 42.02 | 0.2827 | −21 679 | −8849 |
| 42.62 | 0.2995 | −19 571 | −9100 |
| 43.49 | 0.3159 | −19 810 | −9350 |
| 44.43 | 0.3318 | −22 150 | −9648 |
| 44.66 | 0.3471 | −18 663 | −9854 |
| 45.23 | 0.3619 | −17 709 | −10 032 |
| 46.51 | 0.3764 | −16 394 | −10 177 |
| 47.97 | 0.3907 | −15 631 | −10 302 |
| 48.69 | 0.4045 | −15 198 | −10 416 |
| 51.61 | 0.4186 | −14 575 | −10 511 |
| 53.20 | 0.4323 | −14 049 | −10 595 |
| 58.73 | 0.4468 | −13 516 | −10 669 |
| M3 | | | |
| $m_{Cu}$ = 175.48 mg, $m_{Sn}$ = 656.95 mg | | | |
| | | | −610 |
| 24.84 | 0.0150 | −32 246 | −1084 |
| 27.84 | 0.0312 | −31 533 | −1586 |

Table 1 (*Continued*)

| $m_{\mathrm{Au}}$ (mg) | $x_{\mathrm{Au}}$ | $\overline{\Delta H}_{\mathrm{Au}}$ (J/mol) | $\Delta H_{\mathrm{M}}$ (J/mol) |
|---|---|---|---|
| 28.64 | 0.0474 | −31 283 | −2082 |
| 30.12 | 0.0638 | −30 689 | −2576 |
| 32.99 | 0.0812 | −30 191 | −3088 |
| 34.02 | 0.0984 | −29 365 | −3581 |
| 34.66 | 0.1154 | −28 794 | −4054 |
| 36.28 | 0.1324 | −28 392 | −4523 |
| 39.59 | 0.1503 | −27 907 | −5004 |
| 40.10 | 0.1676 | −27 255 | −5459 |
| 41.33 | 0.1848 | −26 649 | −5896 |
| 41.76 | 0.2014 | −25 702 | −6300 |
| 43.40 | 0.2180 | −25 460 | −6698 |
| 45.09 | 0.2345 | −24 968 | −7084 |
| 45.09 | 0.2504 | −23 801 | −7429 |
| 45.60 | 0.2657 | −22 898 | −7746 |
| 47.80 | 0.2812 | −22 988 | −8067 |
| 48.23 | 0.2961 | −21 988 | −8356 |
| 49.25 | 0.3107 | −21 013 | −8619 |
| 55.15 | 0.3264 | −20 474 | −8888 |
| 55.19 | 0.3414 | −19 626 | −9127 |
| 56.19 | 0.3559 | −19 340 | −9355 |
| 58.26 | 0.3704 | −18 470 | −9559 |
| M4 | | | |
| $m_{\mathrm{Cu}}$ = 175.46 mg, $m_{\mathrm{Sn}}$ = 656.00 mg | | | |
| | | | −610 |
| 26.50 | 0.0160 | −32 145 | −1114 |
| 27.14 | 0.0318 | −31 661 | −1605 |
| 28.96 | 0.0482 | −31 283 | −2106 |
| 31.46 | 0.0653 | −31 330 | −2633 |
| 34.70 | 0.0835 | −37 811 | −3318 |
| 35.45 | 0.1014 | −29 797 | −3835 |
| 36.01 | 0.1189 | −30 287 | −4349 |
| 37.20 | 0.1362 | −28 043 | −4815 |
| 37.74 | 0.1531 | −29 570 | −5300 |
| 37.82 | 0.1694 | −26 993 | −5717 |
| 37.98 | 0.1852 | −26 949 | −6120 |
| 38.73 | 0.2006 | −26 130 | −6499 |
| 39.56 | 0.2158 | −26 271 | −6875 |
| 40.19 | 0.2307 | −23 327 | −7187 |
| 40.24 | 0.2450 | −23 961 | −7499 |
| 41.50 | 0.2592 | −22 693 | −7785 |
| 43.08 | 0.2734 | −23 957 | −8095 |
| 45.17 | 0.2877 | −21 944 | −8368 |
| 48.53 | 0.3025 | −15 506 | −8516 |
| 48.54 | 0.3167 | −20 835 | −8766 |
| 48.74 | 0.3303 | −19 386 | −8979 |
| 50.04 | 0.3438 | −19 859 | −9197 |
| 65.86 | 0.3607 | −19 231 | −9456 |
| 67.38 | 0.3771 | −18 892 | −9699 |
| 68.90 | 0.3931 | −17 476 | −9898 |
| M5 | | | |
| $m_{\mathrm{Cu}}$ = 38.90 mg, $m_{\mathrm{Sn}}$ = 630.20 mg | | | |
| | | | 33 |
| 25.77 | 0.0216 | −31 433 | −647 |
| 29.96 | 0.0456 | −30 471 | −1378 |
| 31.74 | 0.0698 | −31 550 | −2142 |
| 33.80 | 0.0942 | −29 789 | −2867 |
| 38.10 | 0.1202 | −19 416 | −3630 |
| 39.23 | 0.1455 | −28 409 | −4342 |
| 40.12 | 0.1699 | −27 928 | −5016 |
| 40.97 | 0.1934 | −26 882 | −5635 |
| 41.19 | 0.2157 | −26 362 | −6209 |
| 42.70 | 0.2376 | −25 855 | −6758 |
| 43.51 | 0.2587 | −24 690 | −7254 |
| 43.81 | 0.2788 | −23 820 | −7702 |
| 43.98 | 0.2979 | −23 023 | −8108 |
| 44.80 | 0.3163 | −22 654 | −8490 |
| 45.86 | 0.3342 | −21 631 | −8834 |
| 46.30 | 0.3514 | −20 806 | −9142 |
| 46.77 | 0.3678 | −19 738 | −9411 |
| 47.69 | 0.3837 | −19 687 | −9670 |
| −9 776 | 0.3991 | −18 639 | −9893 |
| 48.64 | 0.4138 | −18 237 | −10 097 |
| 50.51 | 0.4283 | −17 352 | −10 277 |
| 51.31 | 0.4423 | −16 747 | −10 436 |
| 56.54 | 0.4570 | −16 088 | −10 584 |
| 62.13 | 0.4723 | −15 412 | −10 720 |

Table 2

Partial and integral enthalpies of mixing resulting from calorimetric measurements at different constant ratios of Au:Sn with Cu dropped from 0 to 55 at.% in the ternary system Au–Cu–Sn at 1273 K

| $m_{\mathrm{Cu}}$ (mg) | $x_{\mathrm{Cu}}$ | $\overline{\Delta H}_{\mathrm{Au}}$ (J/mol) | $\Delta H_{\mathrm{M}}$ (J/mol) |
|---|---|---|---|
| M6 | | | |
| $m_{\mathrm{Au}}$ = 1 060.82 mg, $m_{\mathrm{Sn}}$ = 319.86 mg | | | |
| | | | −10 766 |
| 20.84 | 0.0390 | −6887 | −10 615 |
| 20.93 | 0.0752 | −6504 | −10 460 |
| 21.16 | 0.1092 | −6655 | −10 320 |
| 22.92 | 0.1433 | −6293 | −10 166 |
| 23.31 | 0.1753 | −5872 | −10 005 |
| 23.77 | 0.2057 | −5858 | −9853 |
| 24.65 | 0.2348 | −5697 | −9700 |
| 24.98 | 0.2623 | −5531 | −9550 |
| 25.23 | 0.2881 | −5479 | −9408 |
| 25.35 | 0.3123 | −4848 | −9253 |
| 25.45 | 0.3349 | −4733 | −9104 |
| 26.04 | 0.3566 | −4725 | −8961 |
| 26.09 | 0.3770 | −4468 | −8819 |
| 26.26 | 0.3962 | −4146 | −8675 |
| 26.55 | 0.4145 | −3891 | −8530 |
| 27.78 | 0.4325 | −3653 | −8380 |
| 28.59 | 0.4499 | −3451 | −8229 |
| 31.08 | 0.4676 | −3031 | −8062 |
| 31.13 | 0.4843 | −2963 | −7902 |
| 32.38 | 0.5005 | −2521 | −7733 |
| 34.26 | 0.5166 | −2564 | −7566 |
| 34.92 | 0.5320 | −2183 | −7395 |
| 35.97 | 0.5469 | −1818 | −7218 |
| 36.08 | 0.5608 | −1491 | −7041 |
| M7 | | | |
| $m_{\mathrm{Au}}$ = 589.26 mg, $m_{\mathrm{Sn}}$ = 607.57 mg | | | |
| | | | −10 131 |
| 18.49 | 0.0346 | 2921 | −9679 |
| 18.98 | 0.0678 | 2018 | −9277 |
| 20.17 | 0.1006 | 1027 | −8915 |
| 20.37 | 0.1315 | 651 | −8586 |
| 20.79 | 0.1609 | −85 | −8298 |
| 21.08 | 0.1887 | 42 | −8022 |
| 21.10 | 0.2148 | −279 | −7773 |
| 21.26 | 0.2394 | −1134 | −7564 |
| 22.71 | 0.2641 | −1097 | −7355 |
| 22.88 | 0.2874 | −1582 | −7172 |
| 22.95 | 0.3093 | −1786 | −7006 |
| 23.06 | 0.3300 | −1978 | −6856 |
| 23.45 | 0.3498 | −2156 | −6717 |

Table 2 (*Continued*)

| $m_{Cu}$ (mg) | $x_{Cu}$ | $\overline{\Delta H}_{Au}$ (J/mol) | $\Delta H_M$ (J/mol) |
|---|---|---|---|
| 24.42 | 0.3693 | −2304 | −6585 |
| 25.24 | 0.3882 | −2792 | −6471 |
| 25.52 | 0.4062 | −2874 | −6365 |
| 25.62 | 0.4232 | −3027 | −6270 |
| 25.95 | 0.4395 | −3117 | −6181 |
| 29.83 | 0.4571 | −3408 | −6093 |
| 30.02 | 0.4737 | −3726 | −6021 |
| 30.45 | 0.4896 | −3459 | −5944 |
| 31.95 | 0.5053 | −3902 | −5881 |
| 32.01 | 0.5200 | −4300 | −5834 |
| 33.23 | 0.5344 | −3715 | −5770 |
| 34.25 | 0.5484 | −3414 | −5700 |

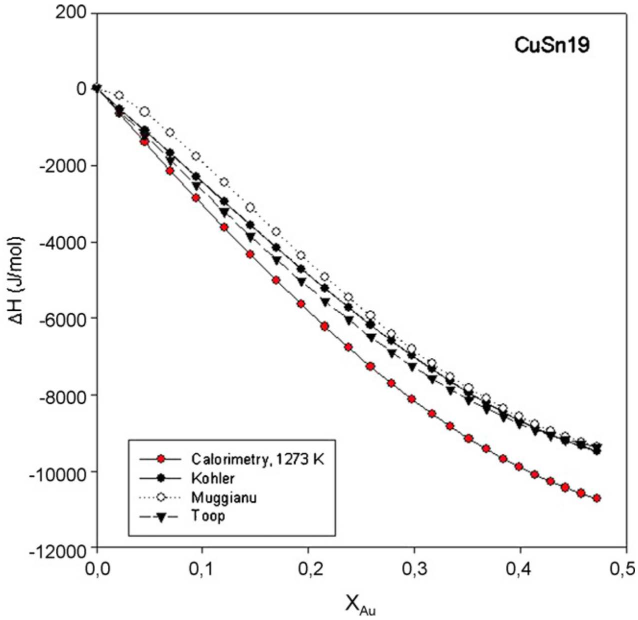

Fig. 4. Integral enthalpy of mixing at the cross section Cu:Sn = 1:9 (M5) compared with the values obtained using the extrapolation models of Kohler, Toop, and Muggianu at 1273 K.

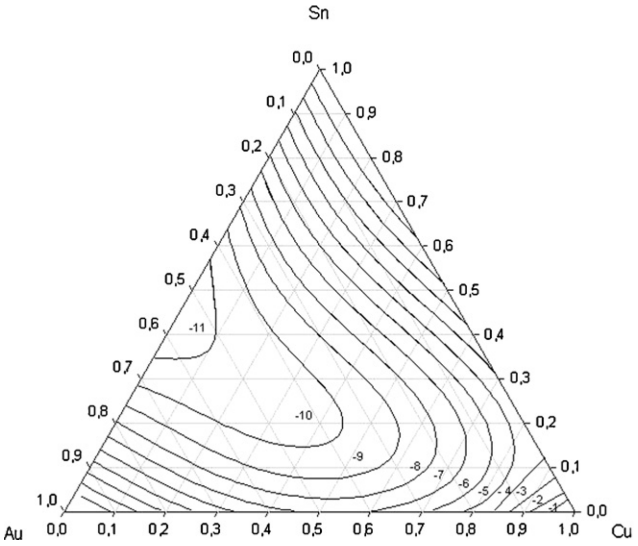

Fig. 5. Iso-enthalpy lines of the ternary Au–Cu–Sn system at 1273 K, obtained experimentally in kJ/mol.

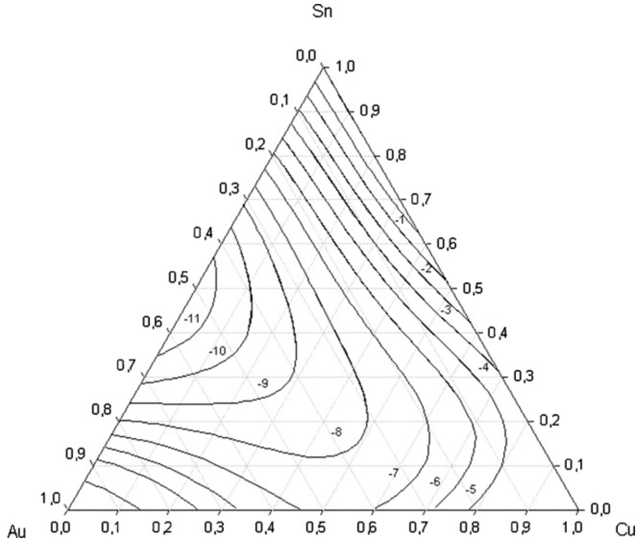

Fig. 6. Iso-enthalpy lines of the ternary Au–Cu–Sn system at 1273 K, obtained by the calculation model of Chou et al. [4] in kJ/mol.

data calculated with the Chou model show a much better agreement with the experimental data than with other calculation or extrapolation models.

## 7. Experimental results

Four measurements have been performed at different cross sections of Cu:Sn (M1–M5) and two measurements have been performed at the cross section Au:Sn (M6, M7). The measured values for the integral enthalpy of mixing are highly negative, up to −11 kJ/mol, which can be seen in Figs. 2 and 3. The results are presented in Tables 1 and 2.

At the Cu:Sn = 1:2 cross section the measurement has been carried out twice (M3, M4) and the reproducibility was excellent, within ±200 J/mol. The experimental errors of these calorimetric measurements are ±5%.

Neither of the calculation models fits well with the experimental data, except at the cross section of Cu:Sn = 1:9, which is shown in Fig. 4. The agreement between the measured Fig. 5 and

Table 3
Binary and ternary interaction parameters related to (Eq. (4))

| Interaction parameter | $\nu$ | J mol$^{-1}$ |
|---|---|---|
| $L^{(\upsilon)}_{\mathrm{Au:Cu}}$ | 0 | −28 474 |
| | 1 | 3373 |
| | 2 | 2339 |
| $L^{(\upsilon)}_{\mathrm{Au:Sn}}$ | 0 | −46 345 |
| | 1 | −9089 |
| | 2 | 6435 |
| $L^{(\upsilon)}_{\mathrm{Cu:Sn}}$ | 0 | −8621 |
| | 1 | −21 742 |
| | 2 | −13 128 |
| $L^{(\upsilon)}_{\mathrm{Au:Cu:Sn}}$ | 0 | −40 103 |
| | 1 | −46 897 |
| | 2 | 53 870 |

the calculated Fig. 6 iso-enthalpy lines, is very good, except at the Au–Sn side, where a rather high discrepancy can be observed.

Table 3 shows the interaction parameter of the binary systems Au–Cu, Au–Sn, and Cu–Sn and the results of the least square fit.

## 8. Conclusion

With the determined data for the integral enthalpy of mixing ternary modelling has been performed. With the obtained set of ternary interaction parameter it is possible to calculate the integral enthalpy of mixing at 1273 K at any concentration of the ternary Au–Cu–Sn system. Various extrapolation models [1–3] and the calculation model given by Chou et al. [4] have been used for the calculation of the integral enthalpy of mixing of the ternary Au–Cu–Sn system.

## Acknowledgement

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