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Pb-free solders: Comparison of different geometrical models in calculating of enthalpy of mixing of In-Sn-Zn ternary system

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"This article is dedicated to our dear Professor Jean Pierre Bros Ex-Professor at the University of Marseille, France"

Abstract
In this paper, the general solution model of Chou has been used to predict the integral enthalpies of mixing of liquid In-Sn-Zn ternary alloys in five selected sections, x_{In}/x_{Sn} = 0.15/0.85, 0.34/0.66, 0.50/0.50, 0.67/0.33 and 0.85/0.15. The other traditional models such as Kohler, Muggianu, Toop and Hillert are also included in calculations. Comparison with literature data was done and showed reasonable agreement with Toop and Hillert asymmetric models.

Keywords: Pb-free solders; Integral enthalpy of mixing; Geometric models; In-Sn-Zn ternary system

1. Introduction
Lead-tin (Pb-Sn) solders have been used in the electronics industry as a primary method for interconnecting electronic components and sub-assemblies for several decades. The wide-spread usage of Pb-Sn solders is due primarily to the combination of low cost and convenient material properties. But, because of the toxic nature of lead, its use is restricted in many applications. So, new lead free solders are required and tin-based multicomponent materials with alloying elements as Ag, Zn, In, Bi, Sb are likely to be most promising. Interest in Sn–Zn based alloys has recently considerably increased again, mainly due to their possible application as high-temperature lead-free solders [1]. One of the concerns about Sn–Zn solders had been that they can easily corrode in high humidity which would clearly be a drawback for the reliability of corresponding solder joints. Various ternary and quaternary additions have been discussed in the past to further improve the properties of binary Sn–Zn solders, among them also the element In. McCormack and Jin [2, 3] reported that the addition of In to Sn–Zn alloys can improve the wetting characteristics of the alloys and lower their melting temperature. Recently, Rechchach et al. [4] have measured the enthalpies of mixing in the liquid ternary In–Sn–Zn alloys over the entire composition at 773 K.
Generally, relatively high investigating temperatures and the evaporation of some metals could make experimental measuring difficult and expensive. That is the main reason for applying thermodynamic predicting methods.
The main purpose of this study is the calculation of the enthalpies of mixing of the liquid phase in the ternary system In-Sn-Zn at 773 K by using the ternary geometric models of Chou [5], Kohler [6], Muggianu et al. [7], Toop [8] and Hillert [9]. The calculated results are compared to the experimental one [4].

2. Literature survey
2.1. Binary In-Sn, In-Zn and Sn-Zn systems
Several calorimetric investigations of the enthalpy of mixing of liquid In-Sn alloys can be found in the literature. They cover the entire composition range and a temperature range from 521 to 1175 K [10-18]. The most recent experimental investigation of the enthalpy of mixing, using direct-reaction calorimetry at 773 K over the entire composition range, was done by Rechchach et al. [4].
The enthalpy of mixing for liquid In-Zn alloys was determined calorimetrically [19-21]. Emf methods were applied to derive the corresponding values [22-24]. All experimental data show positive values for the enthalpy

of mixing. Lee [25] presented a thermodynamic assessment based on the above-mentioned experimental data [19-24]. The calculated and experimental results were in good agreement.
Several calorimetric investigations of the enthalpy of mixing of Sn-Zn liquid alloys at different temperatures are available in the literature [26-30]. Other authors [31-34] have used emf method for this purpose. All experimental data show positive values for the enthalpy of mixing of liquid Sn-Zn alloys. A thermodynamic assessment of the enthalpy of mixing of liquid Sn-Zn alloys were reported [25, 35, 36].

2.2. Ternary In-Sn-Zn system

Enthalpies of mixing of liquid In-Sn-Zn alloys have already been measured by several authors using direct reaction calorimetry [37, 38]. Cui et al. [39], using CALPHAD-type approach have calculated the partial enthalpies of mixing of In, Sn and Zn. Xie et al. [40] used a similar approach for a thermodynamic assessment of the In-Sn-Zn ternary system based on a rather limited number of literature data and observed good agreement with the literature. Recently, Rechchach et al. [4], using a Calvet-type microcalorimeter and a drop calorimetric technique, have measured the partial and the integral enthalpies of mixing of liquid ternary In-Sn-Zn alloys at 773 K along seven sections and over a large composition range. The ternary data of the authors were fitted using the well known Redlich-Kister-Muggianu equation.

3. Theoretical fundamentals

In this work, the investigated system is considered in the order 1-2-3 related to In-Sn-Zn.

3.1 A general solution model of Chou [5]

The method provided by Chou [5] has been proved to be the most reasonable one in all aspects among current geometrical models. This model can not only generalize various kinds of situations, break down the boundary between symmetrical and asymmetrical systems, but can also thoroughly rule out any human interference in the calculation process. The correctness of this model has already been proved theoretically and the accuracy of calculation has also been shown in some practical examples [41, 42]. This model is applied for calculating the enthalpy of mixing of the In-Sn-Zn ternary system.
To apply the Chou's model to the In-Sn-Zn ternary system, it is necessary to calculate the similarity coefficients ξ_{ij} for three binaries which are defined by η_i called the deviation sum of squares:

$$\xi_{12} = \frac{\eta_{\mathrm{I}}}{\eta_{\mathrm{I}} + \eta_{\mathrm{II}}} \quad (1) \qquad \xi_{31} = \frac{\eta_{\mathrm{III}}}{\eta_{\mathrm{III}} + \eta_{\mathrm{I}}} \quad (3)$$

$$\xi_{23} = \frac{\eta_{\mathrm{II}}}{\eta_{\mathrm{II}} + \eta_{\mathrm{III}}} \quad (2)$$

Where

$$\eta_{\mathrm{I}} = \int_0^1 (\Delta_{\mathrm{Mix}} H_{12} - \Delta_{\mathrm{Mix}} H_{13})^2 \mathrm{d}X_1 \quad (4) \qquad \eta_{\mathrm{III}} = \int_0^1 (\Delta_{\mathrm{Mix}} H_{31} - \Delta_{\mathrm{Mix}} H_{32})^2 \mathrm{d}X_3 \quad (6)$$

$$\eta_{\mathrm{II}} = \int_0^1 (\Delta_{\mathrm{Mix}} H_{21} - \Delta_{\mathrm{Mix}} H_{23})^2 \mathrm{d}X_2 \quad (5)$$

and

$$X_{1(12)} = x_1 + x_3\xi_{12} \quad (7) \qquad X_{2(23)} = x_2 + x_1\xi_{23} \quad (8) \qquad X_{3(31)} = x_3 + x_2\xi_{31} \quad (9)$$

The basic equation of the general solution model, derived by Chou, is given as follows:

$$\Delta_{\mathrm{Mix}} H_{123} = x_1x_2 \sum_{\nu} A_{12}^{\nu} (x_1 - x_2)^{\nu} + x_2x_3 \sum_{\nu} A_{23}^{\nu} (x_2 - x_3)^{\nu} + x_3x_1 \sum_{\nu} A_{31}^{\nu} (x_3 - x_1)^{\nu} + x_1x_2x_3 f \quad (10)$$

$\Delta_{\mathrm{Mix}} H_{123}$ is an integral enthalpy of mixing for a ternary 1-2-3 system, x_1, x_2, x_3 are the mole fractions of a ternary alloy, A_{ij}^{ν} are parameters for binary "ij" independent of composition, only relying on temperature, which have been used in the regular type equation:

$$\Delta_{\mathrm{Mix}} H_{ij} = X_iX_j \sum_{\nu} A_{ij}^{\nu} (X_i - X_j)^{\nu} \quad (11)$$

X_{i} and X_{j} indicate the mole fraction of component "i" and "j" in "ij binary system. The function *f* is the ternary interaction coefficient expressed by:

$$f = (2\xi_{12} - 1)\left[A_{12}^2\left((2\xi_{12} - 1)x_3 + 2(x_1 - x_2)\right) + A_{12}^1\right] + (2\xi_{23} - 1)\left[A_{23}^2\left((2\xi_{23} - 1)x_1 + 2(x_2 - x_3)\right) + A_{23}^1\right] + (2\xi_{31} - 1)\left[A_{31}^2\left((2\xi_{31} - 1)x_2 + 2(x_3 - x_1)\right) + A_{31}^1\right] \quad (12)$$

3.2. A traditional Kohler [6], Muggianu et al. [7], Toop [8] and Hillert [9] models

There are several traditional models used to extrapolate the ternary thermodynamic properties based on the three constitutive binary systems. According to Hillert [9], the four traditional models [6-9], generally used due to their simplicity, are classified into two categories: symmetrical (Kohler and Muggianu models) and asymmetrical (Toop and Hillert models). The use of a symmetrical model when an asymmetrical model is more appropriate can often give rise to errors.

The four different extrapolation models [6-9] were used to calculate the enthalpy of mixing in the ternary In-Sn-Zn system which contains three binary systems, and the information of all these binary systems should be known before using the models.

The various predictive extensions from the binary to ternary systems are shown below.

Kholer model [6]:

$$\Delta_{\text{Mix}} H_{123} = (x_{1+}x_2)^2 \Delta_{\text{mix}} H_{12}\left(\frac{x_1}{x_1 + x_2}; \frac{x_2}{x_1 + x_2}\right) + (x_{1+}x_3)^2 \Delta_{\text{mix}} H_{13}\left(\frac{x_1}{x_1 + x_3}; \frac{x_3}{x_1 + x_3}\right) + (x_{2+}x_3)^2 \Delta_{\text{mix}} H_{23}\left(\frac{x_2}{x_2 + x_3}; \frac{x_3}{x_2 + x_3}\right) \quad (13)$$

Muggianu et al. Model [7]:

$$\Delta_{\text{Mix}} H_{123} = \frac{4x_1 x_2}{(1+x_1-x_2)(1+x_2-x_1)} \Delta_{\text{mix}} H_{12}\left(\frac{1+x_1-x_2}{2}; \frac{1+x_2-x_1}{2}\right) + \frac{4x_1 x_3}{(1+x_1-x_3)(1+x_3-x_1)} \Delta_{\text{mix}} H_{13}\left(\frac{1+x_1-x_3}{2}; \frac{1+x_3-x_1}{2}\right) + \frac{4x_2 x_3}{(1+x_2-x_3)(1+x_3-x_2)} \Delta_{\text{mix}} H_{23}\left(\frac{1+x_2-x_3}{2}; \frac{1+x_3-x_2}{2}\right) \quad (14)$$

Toop model [8]:

$$\Delta_{\text{Mix}} H_{123} = (x_{1+}x_2)^2 \Delta_{\text{mix}} H_{12}\left(\frac{x_1}{x_1 + x_2}; \frac{x_2}{x_1 + x_2}\right) + \frac{x_2}{(1 - x_3)} \Delta_{\text{mix}} H_{23}(1 - x_3 ; x_3) + \frac{x_1}{(1 - x_3)} \Delta_{\text{mix}} H_{13}(1 - x_3 ; x_3) \quad (15)$$

Hillert model [9]:

$$\Delta_{\text{Mix}} H_{123} = \frac{x_1 x_2}{\nu_{12}\nu_{21}} \Delta_{\text{Mix}} H_{12}(\nu_{12} ; \nu_{21}) + \frac{x_2}{(1 - x_3)} \Delta_{\text{Mix}} H_{23}(1 - x_3 ; x_3) + \frac{x_1}{(1 - x_3)} \Delta_{\text{Mix}} H_{13}(1 - x_3 ; x_3) \quad (16)$$

Where $\nu_{12} = \frac{1+x_1-x_2}{2}$; $\nu_{21} = \frac{1+x_2-x_1}{2}$

For the calculation of integral enthalpies of mixing of ternary solution according to the Toop [8] and Hillert [9] asymmetrical models one of the three elements has to be chosen as a symmetric component.

4. Results of calculation and discussion

The calculations in the investigated ternary system In-Sn-Zn were carried out along the lines of a constant $x_{\text{In}}/x_{\text{Sn}}$. The five selected cross sections are given in Table 1.

Table 1. The five investigated sections

Section	A	B	C	D	E
$x_{\text{In}}/x_{\text{Sn}}$	0.15/0.85	0.34/0.66	0.50/0.50	0.67/0.33	0.85/0.15

The integral enthalpies of mixing of the three binary systems are taken from Rechchach et al.[4] for In-Sn system at 773 K and from Hultgreen et al.[14] for In-Zn and Sn-Zn systems at 700 and 750 K respectively. Binary regular-solution parameters A_{ij}^{ν} (Eq. (11)) for the binary systems In-Sn, In-Zn and Sn-Zn, used in the calculations are shown in Table 2. It should be noted that:

$\Delta_{\text{Mix}} H_{\text{ij}} = \Delta_{\text{Mix}} H_{\text{ji}}$, $A_{ij}^{\nu} = A_{ji}^{\nu}$, when ν is even and $A_{ij}^{\nu} = -A_{ji}^{\nu}$, when ν is odd.

Table 2. Binary interaction parameters of the integral enthalpy of mixing.

Binary system	A_{ij}^{ν} (J/mol)	Reference
In-Sn	A_{InSn}^{0} = -1488, A_{InSn}^{1} = -1041	[4]
In-Zn	A_{InZn}^{0} = 13095, A_{InZn}^{1} = -2682	[14]
Sn-Zn	A_{SnZn}^{0} = 12728, A_{SnZn}^{1} = -5074	[14]

The plots of integral enthalpies of mixing for the three binary systems at 773 K are shown in Figure 1.

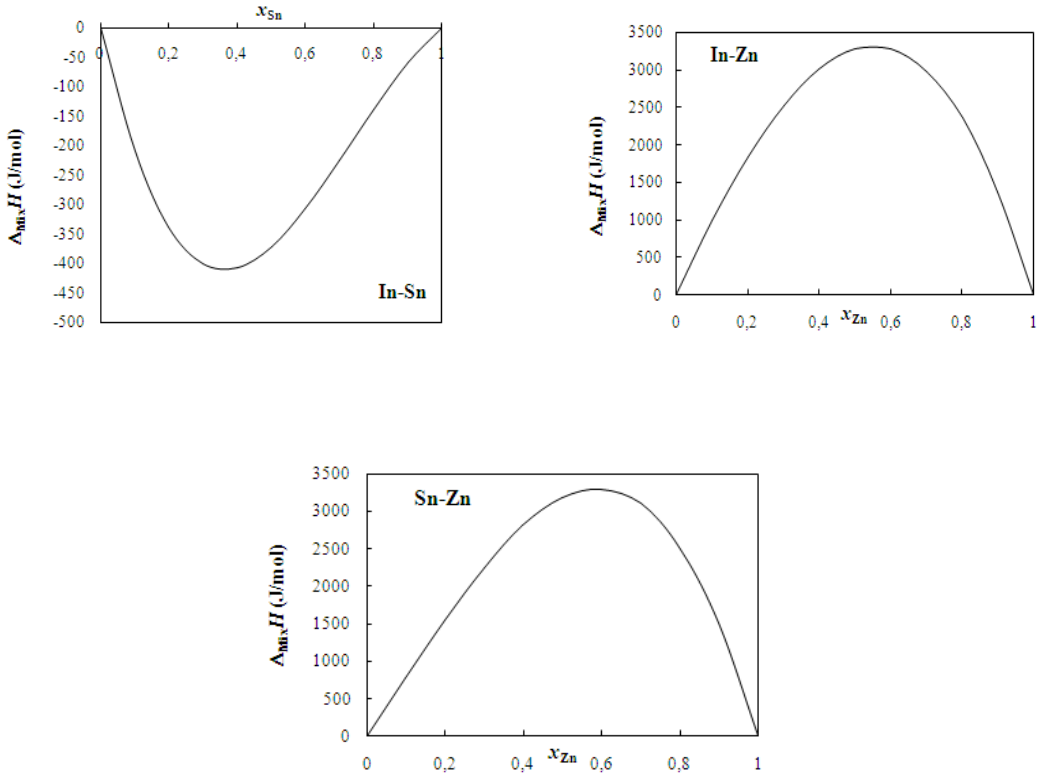

Fig. 1 Integral enthalpies of mixing in the liquid In-Sn (773 K), In-Zn (700 K) and Sn-Zn (750 K) binary systems (Standard states: pure liquid metals).

The enthalpies of mixing are endothermic for In-Zn and Sn-Zn systems with a maximum of about 3200 J/mol at x_{Zn} = 0.60, whereas the In-Sn system has a slight exothermic behaviour with a minimum of - 400 J/mol at x_{In} = 0.58.According to these results, one can conclude that this ternary system should be regarded as an asymmetrical system. The calculated results predicted by Toop or Hillert asymmetric model will be better than those from the Kohler or Muggianu symmetrical model. It has to be mentioned here that, the arrangement of three components to a triangle for an asymmetrical model is very important, because a wrong arrangement will lead to an even worse result than a symmetrical model. In this example, the component "Zn" has been selected as a symmetric point.
Using the Equations (1) to (6) developed by Chou, the deviation sum of squares and the similarity coefficients for the three binaries In–Sn, Sn–Zn and Zn-In are listed in Table 3.

Table 3. Deviation sum squares and similarity coefficients for Chou's model [5] applied on In-Sn-Zn

Deviation sum of squares (J^2/mol^2)	$\eta_I = 7101619.54$		$\eta_{II} = 6914551.51$		$\eta_{III} = 31735.65$	
Interaction of	In-Sn	In-Zn	Sn-In	Sn-Zn	Zn-In	Zn-Sn
Similarity coefficients	$\xi_{In\text{-}Sn} = 0.5067$		$\xi_{Sn\text{-}Zn} = 0.9954$		$\xi_{Zn\text{-}In} = 0.0045$	

From these resulting values (Table 3), one can conclude that the integral enthalpies of mixing of In-Zn and Sn-Zn are more similar to each other than to In-Sn, respectively.
These ξ-values indicate also that this system is an asymmetrical system. In fact, Chou [5] has reported that the three similarity coefficients are not independent. Combining Eqs. (1) to (3) and eliminating η_I, η_{II}, η_{III} the following relation is obtained:

$$(1-\xi_{InSn})(1-\xi_{SnZn})(1-\xi_{ZnIn}) = \xi_{InSn}\,\xi_{SnZn}\,\xi_{ZnIn} \quad (17)$$

In order to check the correction of calculated ξ, the Eq. (17) has been used:

$$(1-\xi_{\mathrm{InSn}})(1-\xi_{\mathrm{SnZn}})(1-\xi_{\mathrm{ZnIn}}) = \xi_{\mathrm{InSn}}\,\xi_{\mathrm{SnZn}}\,\xi_{\mathrm{ZnIn}} = 0.00227 \qquad (18)$$

According to Eq. (17) and reference [5], the above result shows that the calculations of ξ are correct. From Eq. (17), it may be seen that, if any one of the three similarity coefficients approaches zero, then there must be one similarity coefficient which approaches unity. This situation, obtained in this work for our investigated In-Sn-Zn system, represents the common feature of the asymmetrical Toop and Hillert models.

Based on these data, ternary interaction coefficients *f* which enabled determination of integral enthalpies of mixing were calculated according to Eq. (12) for all investigated cross sections in the In-Sn-Zn ternary system, and are given in Table 4.

Table 4. Ternary interaction coefficients, *f* calculated according to Eq. (12) for the In-Sn-Zn system at 773 K (in J/mol)

Mole fraction x_{Zn}	*f*				
	A	B	C	D	E
0	-6303	-6069	-5872	-5662	-5441
0.1	-6686	-6475	-6298	-6110	-5910
0.2	-7069	-6882	-6724	-6557	-6380
0.3	-7452	-7288	-7151	-7004	-6849
0.4	-7835	-7695	-7577	-7451	-7318
0.5	-8219	-8102	-8003	-7899	-7788
0.6	-8602	-8508	-8430	-8346	-8257
0.7	-8985	-8915	-8856	-8793	-8727
0.8	-9368	-9322	-9282	-9240	-9196
0.9	-9752	-9728	-9708	-9688	-9665
1	-10135	-10135	-10135	-10135	-10135

The calculated results for the five models in all the ternary investigated sections (A-E) are listed in Table 5. The experimental results [4] are also included for comparison.

Based on the all calculated results shown in Figure 2, it could be concluded that the comparison between calculated results indicate that the good agreement is obtained between Chou, Toop and Hillert model. In addition, the results obtained by the Toop model show the best agreement with the experimental results.

Additionally, if one takes the root mean square deviation corresponding to experimental results for each traditional model, i.e.

$$S = \frac{1}{N}\sqrt{\sum_{i=1}^{N}(\Delta_{\mathrm{Mix}}H_{\mathrm{the,i}} - \Delta_{\mathrm{Mix}}H_{\mathrm{exp,i}})^2} \qquad (18)$$

Where $\Delta_{\mathrm{Mix}}H_{\mathrm{the,i}}$ and $\Delta_{\mathrm{Mix}}H_{\mathrm{exp,i}}$ represent the integral enthalpies of mixing at a fixed composition "i" for a theoretical model and an experimental results, respectively, while *N* is the total number of investigated alloys. According to the calculation, these S for Toop, Hillert, Chou, Muggianu, and Kohler models are 12.55, 12.79, 12.93, 32.99 and 37.57, respectively. From these S-values, Toop and Hillert asymetric models give the minimum value (12.55 and 12.79 respectively).

Table 5. Integral enthalpies of mixing of In-Sn-Zn system at 773 K calculated by different predicting models and measured by Rechchach et al. [4].

Mole fraction	**Integral enthalpy $\Delta_{\mathrm{Mix}}H$ (J/mol)**					
x_{Zn}	**Kohler**	**Muggianu**	**Chou**	**Toop**	**Hillert**	**Exp. [4]**
Section A : $x_{\mathrm{In}}/x_{\mathrm{Sn}}$ = 0.15/0.85						
0.0	-97	-97	-97	-97	-97	-97
0.1	766	803	734	741	733	750
0.2	1620	1641	1526	1537	1525	1533
0.3	2382	2368	2229	2241	2228	2215
0.4	2980	2933	2790	2802	2788	2753
0.5	3352	3285	3154	3165	3153	3100
0.6	3441	3371	3266	3274	3265	3205

0.7	3196	3139	3067	3072	3066	3011
0.8	2570	2536	2498	2500	2497	2457
0.9	1519	1508	1496	1497	1496	1477
1.0	0	0	0	0	0	0
Section B : x_{In}/ x_{Sn} = 0.34/0.66						
0.0	-259	-259	-259	-259	-259	-259
0.1	695	771	653	658	652	645
0.2	1618	1685	1487	1496	1486	1451
0.3	2433	2446	2206	2215	2204	2135
0.4	3062	3015	2767	2776	2765	2666
0.5	3442	3352	3125	3133	3123	3006
0.6	3519	3414	3231	3237	3229	3110
0.7	3247	3157	3031	3035	3030	2924
0.8	2591	2534	2467	2469	2467	2390
0.9	1517	1498	1479	1479	1479	1440
1.0	0	0	0	0	0	0
Section C : x_{In}/ x_{Sn} = 0.50/0.50						
0.0	-372	-372	-372	-372	-372	-372
0.1	653	737	610	609	609	564
0.2	1617	1693	1478	1476.1248	1476	1390
0.3	2448	2469	2207	2204.6388	2205	2085
0.4	3079	3037	2764	2762.4048	2762	2620
0.5	3452	3364	3114	3111.75	3112	2961
0.6	3519	3413	3211	3209.0928	3209	3064
0.7	3240	3145	3006	3004.9428	3005	2879
0.8	2578	2519	2444	2443.9008	2444	2351
0.9	1507	1487	1465	1464.6588	1465	1414
1.0	0	0	0	0	0	0
Section D : x_{In}/ x_{Sn} = 0.67/0.33						
0.0	-407	-407	-407	-407	-407	-407
0.1	674	741	631	624	631	550
0.2	1655	1707	1521	1510	1520	1394
0.3	2470	2476	2248	2235	2247	2100
0.4	3072	3029	2792	2779	2790	2638
0.5	3418	3340	3122	3111	3120	2973
0.6	3470	3380	3202	3193	3201	3065
0.7	3188	3109	2987	2981	2986	2867
0.8	2539	2489	2423	2420	2423	2328
0.9	1487	1470	1451	1450	1451	1393
1.0	0	0	0	0	0	0
Section E : x_{In}/ x_{Sn} = 0.85/0.15						
0.0	-283	-283	-283	-283	-283	-283
0.1	818	841	780	772	779	690
0.2	1762	1766	1661	1649	1661	1544
0.3	2510	2490	2361	2347	2360	2250
0.4	3043	3003	2869	2854	2868	2774
0.5	3339	3287	3163	3150	3162	3083
0.6	3367	3313	3212	3203	3212	3139
0.7	3091	3047	2977	2970	2976	2902
0.8	2470	2443	2405	2402	2405	2330
0.9	1457	1448	1437	1436	1437	1378
1.0	0	0	0	0	0	0

Therefore, according to the S-values, one can confirm that the results obtained by Toop model are close to the experimental one and the investigated In-Sn-Zn system is an asymmetric system.

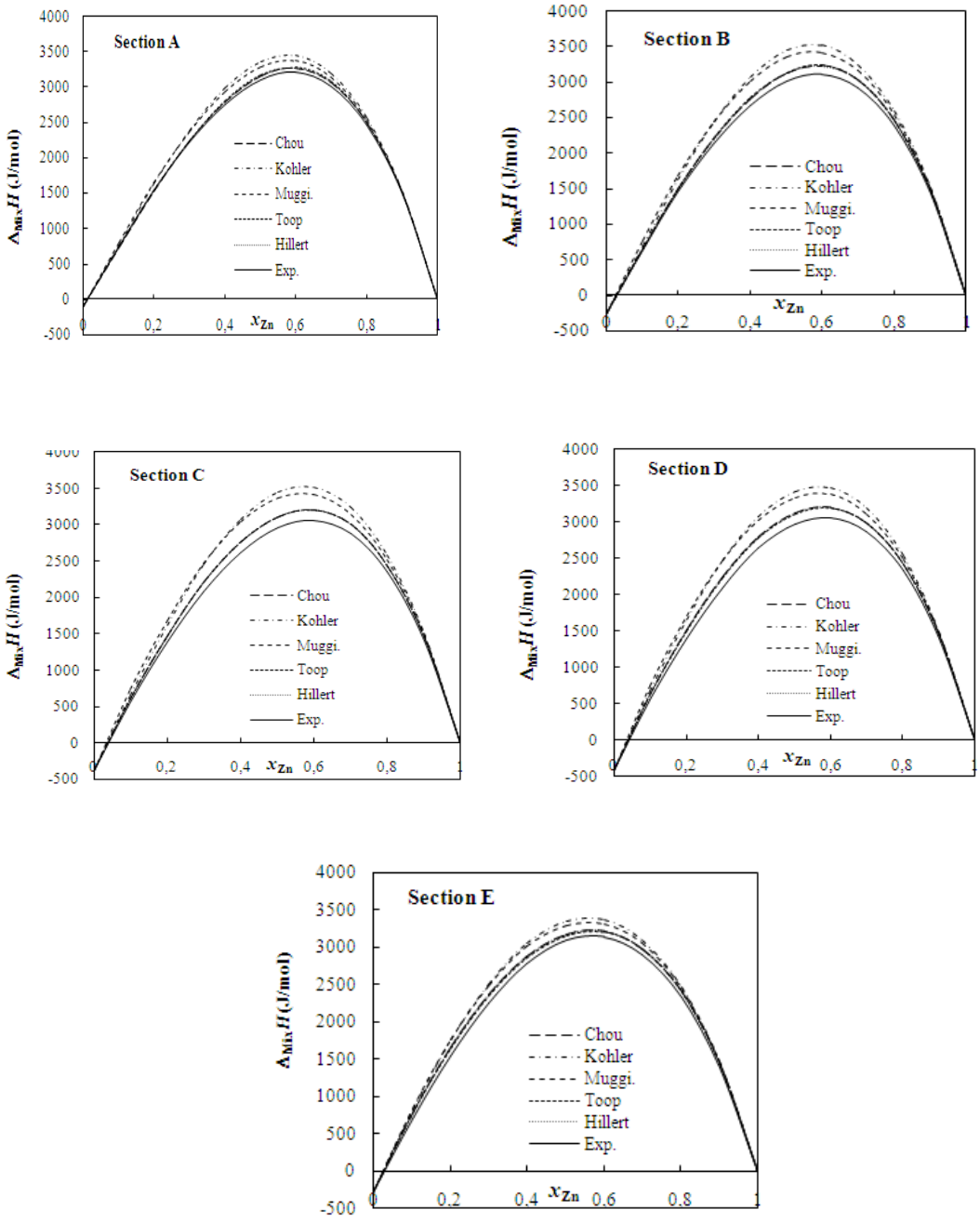

Fig. 2 Integral enthalpies of mixing in the liquid In-Sn-Zn alloys at 773 K using the different models, together with the experimental data [4] (Standard states: pure liquid metals).

Conclusion

The integral enthalpies of mixing of the ternary In-Sn-Zn system were calculated by using the Chou, Kohler, Muggianu, Toop and Hillert models and compared with the experimental results. Among all the traditional predicting models, it can be concluded that Toop and Hillert models show the best agreement with the experimental data. This conclusion can be explained by the fact that the asymmetric models are the closest to the obtained geometric interpretation for the In-Sn-Zn system.

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