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Prediction of surface tension, viscosity and molar volume of Ag-Bi-Sn liquid alloys at different temperatures

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ABSTRACT

The surface tensions, viscosities and molar volumes of liquid Ag-Bi-Sn alloys were calculated and presented in this paper. These physical properties have been estimated using the Kohler's model for surface tension and molar volume and Seetharaman's model for viscosity. The calculations were done along three selected sections with x_{Sn}/x_{Bi} = 1/2, 1/1 and 2/1 at different temperatures from 873 to 1273 K. The results show that up to 40 at. % Ag, no influence of the surface tension and no significant dependence on temperature has been noted. With increasing temperature the viscosity decreases smoothly. Both surface tension and viscosity increase significantly at high Ag-concentrations. The molar volume decreases linearly with increasing Ag-compositions and the calculated values are similar to those of additive molar volumes.

1. Introduction

Sn–Pb solders have been the most important material for the interconnection in electronic components. They are used because of their low cost and good physical, chemical and mechanical properties. However, due to environmental and health concerns alternative solder alloys are needed [1]. So, the development of a good Pb-free solder which is environmentally friendly materials is a great challenge.

New solders must fulfill several requirements and physico-chemical properties like good wettability, high corrosion resistance and melting point which must be closer to the conventional Sn-Pb solders. Researches in the field of the lead-free solders are thus important and the elements which are possible candidates for solders in Sn-base alloys are: Bi, Ni, In, Ag, Zn, Sb, etc.

Among several pure metals with good conductivity, it is found that Ag is a suitable substrate to form only one layer inter-metallic compounds (Ag_3Sn) after chemical reactions with most Sn-based solders [2-4]. It is reported that the simple interfacial reaction between Ag and the solders is beneficial to the soldering joint reliability [2-4]. The Ag-Bi-Sn alloys are particularly interesting because of their potential Pb-free solder applications.

As the surface properties play a key role in joining process, development of new solders requires data on thermo-physical properties such us surface tension and viscosity which can be obtained experimentally or by numerical modeling, if a reliable data for pure components and simple subsystems are present. Moreover, numerical modeling is sometimes preferred because of growing complexity and cost experiments, especially for the multicomponent systems (ternary, quaternary…).

If the thermodynamic properties (enthalpy of mixing, free enthalpy of mixing and phase diagram) of the liquid Ag-Bi-Sn alloys have been widely studied [5-10], however, few investigations on the physical properties such us surface tension, viscosity, density have been reported on the literature [11-14]. Indeed, Moser et al. [11] have measured the surface tension of liquid Ag-Bi-Sn alloys adding small compositions of Bi to eutectic Ag-Sn. They have reported that the linear dependencies of surface tensions on temperature were observed. Later, Terzieff et al. [12] have deduced the surface tension of the liquid phase from the experimental thermodynamic activity

coefficients and calculated the viscosity. Fima and Kucharski [13] have also measured the surface tension of the same system and observed a curvilinear dependence of surface tension with temperature for all of the compositions, except for those of a low Ag content. Very recently, the surface tension of the liquid Ag-Bi-Sn ternary alloys has been measured with the sessile drop method by Hindler [14] along three cross sections Ag:Bi = 2:1, 1:1 and 1:2. No influence of silver on the surface tension has been observed. With increasing Bi compositions, the surface tension decreases slightly.

Moreover, reliable viscosity data as a function of temperature metallic melts are important for modeling and designing in metallurgical processes. For example, the rate of impurities transferring from metallic melts to slag is affected by the viscosity of metallic melts.

Although a number of recent experimental measurements have been carried out for thermophysical properties in the liquid sub-binary Ag-Bi [15, 16], Ag-Sn [17, 18] and Bi-Sn [19, 20] systems of Ag-Bi-Sn, the amount of avaible data is still scare especially for ternary and multicomponent systems [11-14].

In our recent paper [21], the surface tensions of the Ag-Bi-Sn ternary alloys at 873 K are calculated from the surface tensions of the Ag-Bi, Ag-Sn and Bi-Sn sub-binary systems by using Kohler [22], Toop [23] and Hillert [24] geometric models, and by using the Butler's equation [25]. Except for the asymmetric Toop and Hillert models, the agreement between the calculated results and experimental data is quite good using Kohler and Butler's models. In this work, the surface tension and the viscosity for ternary Ag-Bi-Sn liquid alloys were calculated over temperature ranges (873-1273 K) using the Kohler (based on the results obtained in our previous paper [21]) and Seetharaman and Sichen [28] models, respectively. The SURDAT database [26] which contains data of surface tension and molar volume in an extensive range of temperatures and concentrations has been used. The choice of the temperature range 823-1273K is based on the fact to compare our calculations with the experimental work reported by Fima and Kucharski [13].

2. Theoretical fundamentals

2.1. Surface tensions calculation of liquid Ag-Bi-Sn system

2.1.1. Excess surface tension of sub-binary systems

The contributions for three sub-binary systems of the Ag-Bi-Sn ternary alloys are described by using the Redlich-Kister (R-K) [27] Polynomial:

$$\sigma^E = X_1 X_2 \sum_{i=0}^{n} A_i (X_1 - X_2)^i \qquad (1)$$

Where A_i is the coefficient of the R-K polynomial. The coefficients of R-K polynomial for excess surface tensions of the three sub-binary alloys as function the temperature are shown in Table1.

Table 1: Optimized parameters A_i (mN/m) for excess surface tensions of the Ag-Bi, Ag-Sn and Bi-Sn sub-binary alloys as function of the temperature [26] using in the present work.

System	A_0	A_1	A_2
Ag-Bi	-1492.9+0.76147T-0.0001216T^2	1738.4-1.37566T+0.000404T^2	-1904.2+2.33459T-0.0009596T^2
Ag-Sn	-1350.7+0.93116T-0.0001902T^2	1300.7-1.39459T+0.0004202T^2	-1063.5+1.91129T-0.0006179T^2
Bi-Sn	-305.5+0.29948T-0.0000947T^2	247.9-0.29995T+0.0001027T^2	-412.4 + 0.65851T - 0.0002686T^2

2.1.2. Calculation of surface tension of liquid phase in the Ag-Bi-Sn System

The excess surface tension σ^E of melts is defined as the difference between the true surface tension and the corresponding value calculated from the ideal solution model. Using Kohler's model [22], the surface tension of ternary alloy systems is given by:

$$\sigma^E = (x_1 + x_2)^2 (\sigma_{12}^E)_{\frac{x_1}{x_2}} + (x_1 + x_3)^2 (\sigma_{13}^E)_{\frac{x_1}{x_3}} + (x_2 + x_3)^2 (\sigma_{23}^E)_{\frac{x_2}{x_3}} \qquad (2)$$

Where $\sigma_{i,j}^{E}$ denotes the excess surface tension of the binary system i-j for the ratio $X_i/X_j=x_i/x_j$, x_i and X_i represent the molar fraction of a component i in the ternary and binary systems, respectively.

2.2. Viscosity calculation of liquid Ag-Bi-Sn system

Seetharaman and Sichen [28] have proposed the following equation for the viscosity for liquid alloys.

$$\eta = \frac{hN_0\rho}{M}\exp\left(\frac{\Delta G^*}{RT}\right) \quad (3)$$

$$\rho = \sum x_i\rho_i \quad , \; M = \sum x_i M_i$$

Where ρ and ρ_i are the densities of liquid alloy and pure component i, respectively. M and M_i are atomic weights of the liquid alloy and pure component i, respectively, h is Planck's constant. N_0 is Avogadro's number. ΔG^* is the Gibbs energy of activation for viscosity, which can be expressed in a multi-component system as follows.

$$\Delta G^* = \sum x_i \Delta G_i^* + 3RT\sum x_i x_j + RT\sum x_i \ln x_i + \Delta G^E(T, x_B) \quad (4)$$

Where ΔG_i^* is the Gibbs energy of activation of the viscous flow in pure component *i*

$$\Delta G_i^* = RT\ln\left(\frac{\eta_i M_i}{hN_0\rho_i}\right)$$

With η_i is the viscosity of the pure component i (Pa.s) and R = 8.314 J.mol^{-1}K^{-1}, the universal gaz constant. T is the absolute temperature (K), and $\Delta G^E(T, x_B)$ is the excess energy of alloy in the liquid phase, which can also be obtained from the thermodynamic database.

Table 2: Data on viscosity, density and atomic mass of the pure components

Component	η_i (mPa.s)	Ref.	ρ_i (g/cm^3)	Ref.	M_i (g/mol)
Ag	0.5976exp(19137/*RT*)	[29]	10.8-0.00071T	[32]	107.8680
Bi	0.4458exp(6450/*RT*)	[30]	10.73-0.0012T	[33]	208.9804
Sn	0.3847exp(7512/*RT*)	[31]	7.118-0.00051T	[33]	118.6900

ΔG^{E} has been calculated using the kohler model as follows:

$$\Delta \mathrm{G^E} = (\mathrm{x_1}+\mathrm{x_2})^2(\Delta G_{12}^{\mathrm{E}})_{\frac{\mathrm{x_1}}{\mathrm{x_2}}} + (\mathrm{x_1}+\mathrm{x_3})^2(\Delta G_{13}^{\mathrm{E}})_{\frac{\mathrm{x_1}}{\mathrm{x_3}}} + (\mathrm{x_2}+\mathrm{x_3})^2(\Delta G_{23}^{\mathrm{E}})_{\frac{\mathrm{x_2}}{\mathrm{x_3}}} \quad (5)$$

Where $\Delta G_{\mathrm{i,j}}^{\mathrm{E}}$ is the free enthalpy of liquid binary ij alloys wich can be obtaind by the know polynomial of Redlich-Kister (R-K) [27]. The Coefficients of the R-K polynomial for excess energy of the Ag-Bi, Ag-Sn and Bi-Sn are shown in table3.

Table 3: Interaction parameters (J/mol) for the Ag–Bi-Sn system

System	L^0	L^1	L^2	Ref.
Ag-Bi	3340.81+39.16749*T*-5.969876*T*Ln*T*	-5485.45-1.07133*T*	-3055.34+1.7749*T*	[34]
Ag-Sn	-5146.7-5.0103*T*	-15799.3+3.3208*T*	-6687.5	[35]
Bi-Sn	490+0.966*T*	-30-0.235*T*	0	[36]

2.3. Molar volume calculation of liquid Ag-Bi-Sn system

2.3.1. Equation for the excess molar volume of sub-binary systems

The composition dependence of the molar volume of mixtures can be represented in terms of the excess molar volume V^E defined as:

$$V^E = V - V^i \quad (6)$$

Where V is the molar volume of a binary liquid system, V^i is the molar volume of the ideal solution, and V^ican be obtained from the following equation:

$$V^i = X_1V_1 + X_2V_2 \quad (7)$$

Where X_i is the molar fraction of a component i, and V_i is the molar volume of pure liquid i. The molar volumes are shown in table 4.

Table 4: Molar volume of the pure components.

Component	V_i (m^3/mol)	Reference
Ag	$11.6.10^{-6}$(1.0+0.000098(*T*-1234))	[37]
Bi	$20.80.10^{-6}$ (1.0+0.000117(*T*-544.1))	[38]
Sn	$17.0.10^{-6}$(1.0+0.000087(*T*-504.99))	[38]

In this work, the excess molar volume values of the sub-binary systems $V_{i,j}^E$ are taken from previous work of Kucharski and Fima [33].These values will be used to calculate the excess molar volumes of the ternary Ag-Bi-Sn alloys.

2.3.2. Calculation of molar volume of liquid phase in the Ag-Bi-Sn systems using Kohler's model

The excess molar volume V^E of melts is defined as the difference between the true volume molar and the corresponding value calculated from the ideal solution model (Eq. 6). Using Kohler's model [22], the excess molar volume of ternary alloys is given by:

$$V^E = (x_1 + x_2)^2 (V_{12}^E)_{\frac{X_1}{X_2}} + (x_1 + x_3)^2 (V_{13}^E)_{\frac{X_1}{X_3}} + (x_2 + x_3)^2 (V_{23}^E)_{\frac{X_2}{X_3}} \quad (8)$$

Where $V_{i,j}^E$ denotes the excess molar volume of the binary system i-j for the ratio $X_i/X_j=x_i/x_j$. x_i and X_i represent the molar fraction of a component i in the ternary and binary systems, respectively.

2. Experimental detail

3.1. Surface tension in the liquid Ag-Bi-Sn ternary alloys

The physic-chemical properties such as surface tension and viscosity calculations of ternary Ag-Bi-Sn alloys were undertaken at three pseudo-binary sections (Fig.1 and Table 5) in the whole range of concentrations and a wide range of temperatures from 873 to 1273 K.

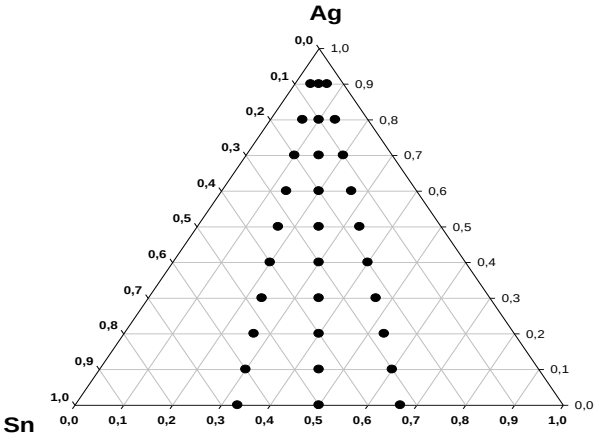

Figure 1: Gibb's triangle with three pseudo-binary sections x_{Sn}/x_{Bi} = 0.5, 1 and 2

Table 5: Compositions and temperatures studied alloys

x_{Sn}/x_{Bi}	x_{Ag}	**Temperature range (K)**
0.5	0.1 - 0.9	873-1273
1	0.1 - 0.9	873-1273
2	0.1 - 0.9	873-1273

All the calculated surface tensions at different temperatures using Kohler's model are shown in Table 6.

Table 6: Calculated surface tensions (N/m) Ag-Bi-Sn ternary alloys at different temperatures

x_{Ag}	**873*K***	**973*K***	**1073*K***	**1173*K***	**1273*K***
Section A : x_{Sn}/x_{Bi} = 1					
0.1	0.4468	0.4506	0.4545	0.4587	0.4625
0.2	0.4279	0.4271	0.4257	0.4237	0.4200
0.3	0.4228	0.4247	0.4259	0.4263	0.4243
0.4	0.4222	0.4277	0.4320	0.4351	0.4347
0.5	0.4245	0.4356	0.4448	0.4522	0.4550
0.6	0.4546	0.4733	0.4899	0.5045	0.5138
0.7	0.5434	0.5758	0.6068	0.6365	0.6612
0.8	0.6607	0.7181	0.7765	0.8359	0.8919
0.9	0.7544	0.8276	0.9046	0.9853	1.0649
Section B : x_{Sn}/x_{Bi} = 0.5					
0.1	0.4650	0.4808	0.4976	0.5157	0.5343
0.2	0.4111	0.4132	0.4147	0.4156	0.4148
0.3	0.4068	0.4093	0.4108	0.4114	0.4094
0.4	0.4129	0.4187	0.4232	0.4263	0.4259
0.5	0.4304	0.4390	0.4456	0.4500	0.4497
0.6	0.4513	0.4634	0.4732	0.4807	0.4826
0.7	0.5050	0.5258	0.5455	0.5642	0.5779
0.8	0.6183	0.6639	0.7111	0.7601	0.8065
0.9	0.7397	0.8173	0.8991	0.9849	1.0701
Section C : x_{Sn}/x_{Bi} = 2					
0.1	0.4552	0.4522	0.4489	0.4453	0.4408
0.2	0.4529	0.4510	0.4485	0.4455	0.4409
0.3	0.4464	0.4477	0.4483	0.4480	0.4454
0.4	0.4356	0.4416	0.4464	0.4500	0.4504
0.5	0.4385	0.4525	0.4652	0.4765	0.4838
0.6	0.4870	0.5150	0.5417	0.5672	0.5881
0.7	0.5831	0.6310	0.6780	0.7240	0.7653
0.8	0.6881	0.7544	0.8210	0.8878	0.9506
0.9	0.7752	0.8356	0.8989	0.9649	1.0288

As an example, Fig. 2 shows the calculated results of surface tensions along section C (addition of silver ($0.1 \leq x_{Ag} \leq 0.9$) to $x_{Sn}/x_{Bi} = 2$). The increase in concentration of Ag (up to 40 at. % Ag) does not influence the surface tension of the Ag-Bi-Sn alloys and no significant dependence on temperature has been noted (Fig. 2). Moreover, for concentrations of Ag higher than 40 at. % the surface tension increases significantly with Ag content and temperature. These results may be explained by the interaction parameters used in our calculation (**See Table 1**).

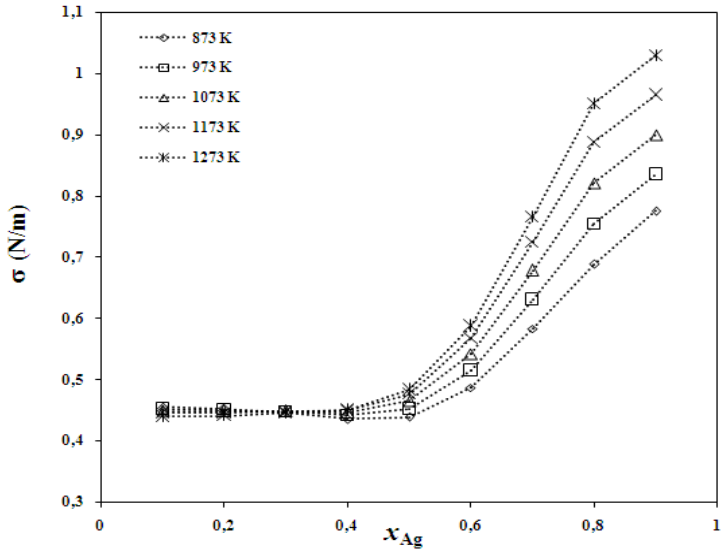

Figure 2: Surface tension isotherms versus molar fraction of Ag for x_{Sn}/x_{Bi}=2/1

In order to have more information in the region $0 < x_{Ag} < 0.4$, we have plotted in Figure 3 the variation of the surface tension in function of the molar fraction of silver at different temperatures. As can be seen in Figure 3, the surface tension does not vary significantly with the composition of silver, but it is clear that this thermo-physical property decrease with increasing temperature, however for the alloys with $x_{Ag} > 0.3$, the phenomenon is reversed.

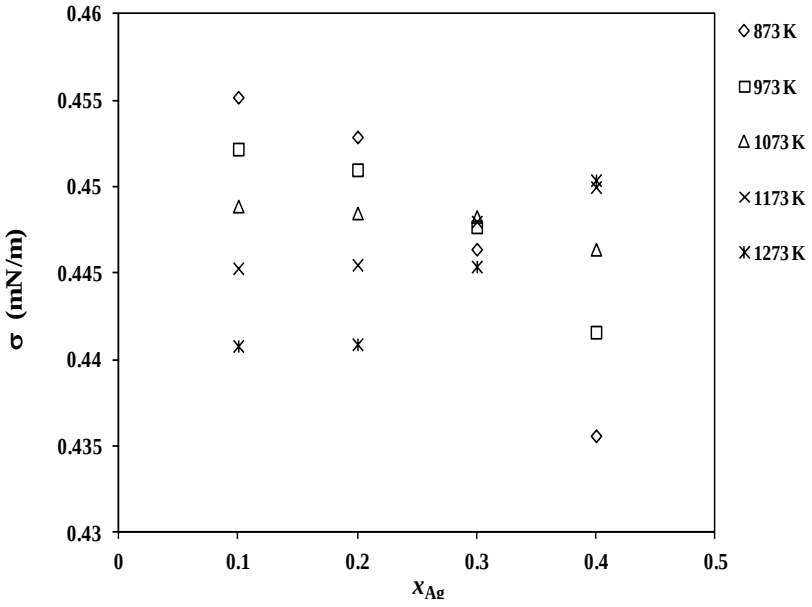

Figure 3: Surface tension isotherms vs. molar fraction of Ag for x_{Sn}/x_{Bi}=2/1 at lower Ag-content

Figure 4 shows the linear temperature dependence of surface tension for the Sn-rich section (x_{Sn}/x_{Bi}=2/1) at different compositions of silver. A linear dependence on temperature is noted. The same behavior was observed for the two other sections A ($x_{Sn}/x_{Bi} = 1$) and B ($x_{Sn}/x_{Bi} = 0.5$).

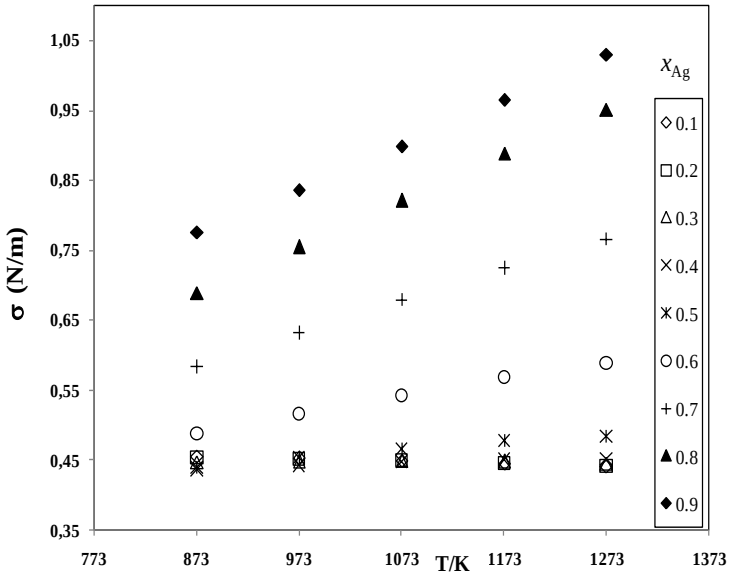

Fig. 4 Temperature dependence of surface tension for a fixed ratio Sn/Bi = 2/1

The calculated values of the surface tension for Ag-Bi-Sn alloys at 1173 K were compared with the experimental one reported by Fima and Kucharski [13] at the same temperature. From Figure 5, one can conclude that the calculated surface tension values are higher than those obtained experimentally [13] for the alloys with $x_{Ag} > 0.5$..

This difference is probably due to the Redlich-Kister parameters (**See Table 1)** used for excess surface tensions of the Ag-Bi, Ag-Sn and Bi-Sn sub-binary alloys in our calculation.

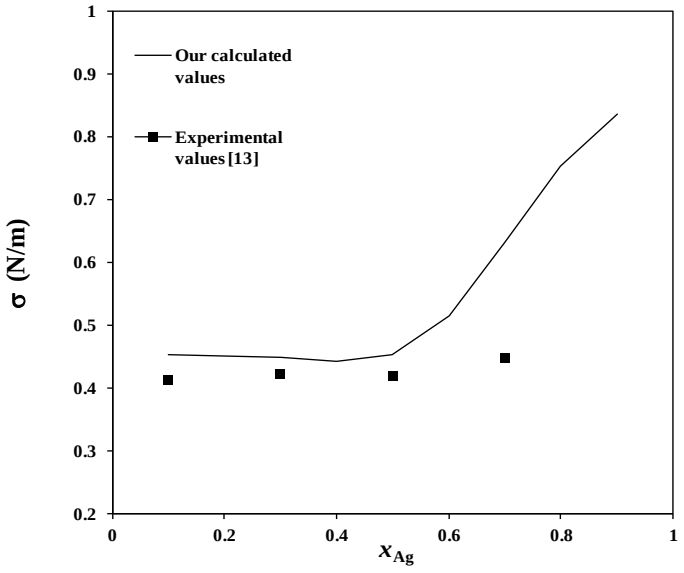

Figure 5: Comparison between the surface tension calculated in this work (lines) and experimental values [13] (symbols) at 873 K for a fixed ratio Sn/Bi = 2/1

3.2. Viscosity in the liquid Ag-Bi-Sn alloys

In spite of the importance of the liquid's viscosity in practical metallurgical process, the experimental information of the liquid alloys, especially for multicomponent systems (ternary, quaternary…), is still not abundant. This is probably due to high reactivity of metallic liquid, experimental difficulties, and time consuming, particularly at high temperatures. It is well known that the adding of a third element into a binary alloy will change the liquid structure in the melt which should be responsible for the variation in the viscosity.

In this work, the calculation of viscosities for liquid Ag-Bi-Sn alloys was done using the Seetharaman and Sichen [28] model along three cross sections Sn:Bi =1:1, 1:2 and 2:1 at different temperatures. The calculated results are summarized in Table 7. To the best knowledge of the authors no experimental data for the viscosity of this system is available from the literature.

Table 7: calculated viscosity of the Ag-Bi-Sn ternary alloys at different temperatures by using equation the model of Seetharaman and Sichen (mPa.S).

x_{Ag}	873K	973K	1073K	1173K	1273K
Section A : x_{Sn}/x_{Bi} = 1					
0.1	1.2358	1.0883	0.9818	0.9016	0.8376
0.2	1.4754	1.2743	1.1316	1.0250	0.9415
0.3	1.7421	1.4840	1.3032	1.1690	1.0650
0.4	2.0304	1.7126	1.4920	1.3290	1.2038
0.5	2.3630	1.9767	1.7108	1.5150	1.3657
0.6	2.7882	2.3113	1.9861	1.7479	1.5678
0.7	3.3879	2.7749	2.3621	2.0620	1.8375
0.8	4.3025	3.4648	2.9101	2.5116	2.2176
0.9	5.7885	4.5523	3.7521	3.1877	2.7784
Section B : x_{Sn}/x_{Bi} = 0.5					
0.1	1.1991	1.0571	0.9543	0.8764	0.8147
0.2	1.3974	1.2062	1.0702	0.9684	0.8889
0.3	1.6253	1.3809	1.2096	1.0823	0.9842
0.4	1.8877	1.5844	1.3742	1.2191	1.1006
0.5	2.2120	1.8371	1.5801	1.3915	1,2487
0.6	2.6464	2.1745	1.8546	1.6215	1.4464
0.7	3.2714	2.6547	2.2421	1.9440	1.7224
0.8	4.2242	3.3740	2.8143	2.4147	2.1214
0.9	5.7551	4.5026	3.6950	3.1275	2.7174
Section C : x_{Sn}/x_{Bi} = 2					
0.1	1.2683	1.1162	1.0068	0.9246	0.8588
0.2	1.5559	1.3449	1.1956	1.0843	0.9966
0.3	1.8782	1.6034	1.4110	1.2684	1.1574
0.4	2.2169	1.8763	1.6398	1.4650	1.3302
0.5	2.5810	2.1693	1.8855	1.6763	1.5161
0.6	3.0106	2.5108	2.1691	1.9180	1.7272
0.7	3.5832	2.9552	2.5305	2.2205	1.9874
0.8	4.4402	3.5997	3.0406	2.6372	2.3382
0.9	5.8475	4.6204	3.8233	3.2592	2.8489

Figure 6 illustrates the calculated values of viscosities for the section x_{Sn}/x_{Bi} = 2/1 as an example. It can be seen that with the increase of temperature, the viscosity decreases smoothly, but increases with increasing Ag-concentrations followed by a significant increase towards the Ag-rich side, similar to the trend observed in binary Sn-Ag previously reported by Gebhardt et al. [39]. The same behavior was observed for the two other sections A (x_{Sn}/x_{Bi} = 1) and B (x_{Sn}/x_{Bi} = 0.5).

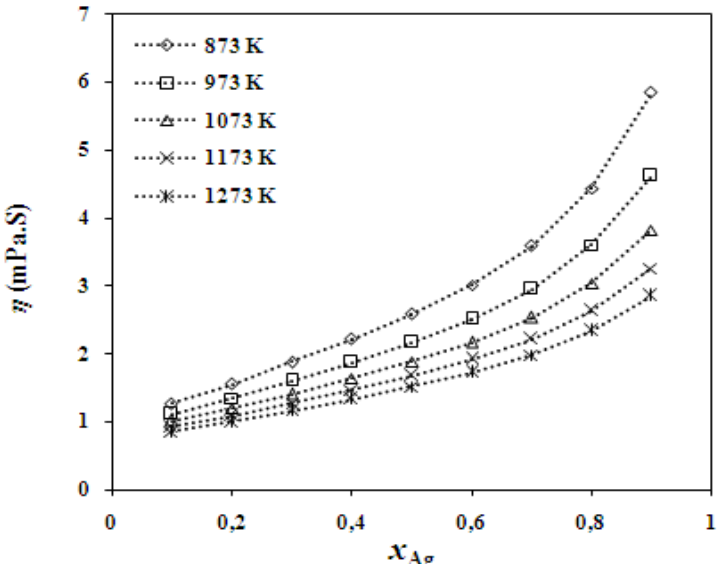

Figure 6: Calculated viscosities isotherms versus molar fraction of Ag for x_{Sn}/x_{Bi}=2/1

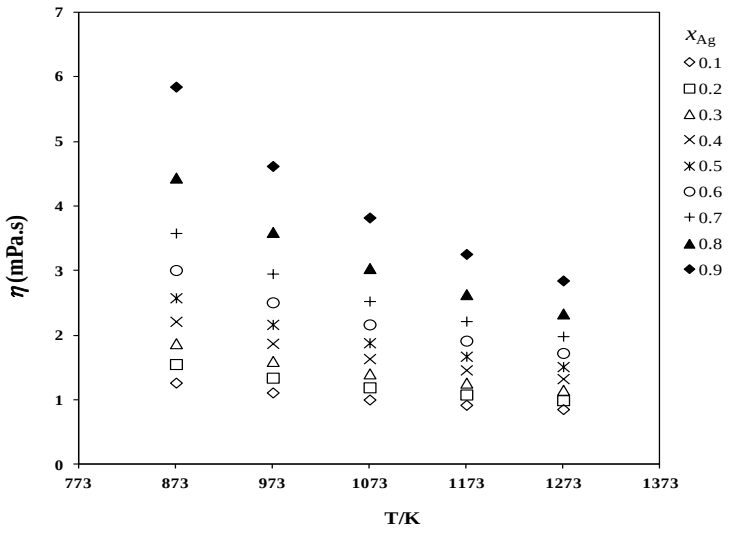

Figure 7: Temperature dependence of viscosity of Ag-Bi-Sn liquid alloys for a section x_{Sn}/x_{Bi} = 2/1

The temperature dependence of viscosity for the Sn-rich section (x_{Sn}/x_{Bi}=2/1) at different compositions of silver is reported in Figure 7. It is clear that the viscosity decreases with increasing temperature. This dependence becomes more significant for the silver rich alloys.

3.3. Molar volume of the liquid Ag-Bi-Sn ternary alloys

The molar volumes were calculated applying Eqs. (6)-(8). The values obtained are shown in Table 8.

Figure 8 illustrates a calculated molar volume isotherms as a function of molar fraction of Ag for the equimolar section (x_{Sn}/x_{Bi} = 1). The calculated molar volumes of ternary Ag-Bi-Sn alloys decrease linearly with increasing

Ag amount. Figure 9 shows a comparison of our calculated values at 1173 K at the same section with a values of Fima and Kucharski [13] deduced from a density measurements. A good agreement is noted.

Table 8: calculated molar volume (cm^3/mol) Ag-Bi-Sn ternary alloys at different temperatures

x_{Ag}	873*K*	973*K*	1073*K*	1173*K*	1273*K*
Section A : x_{Sn}/x_{Bi} = 1					
0.1	19.6461	19.8706	20.1033	20.3375	20.5615
0.2	18.7335	18.9518	19.1780	19.4047	19.6264
0.3	17.8272	18.0363	18.2525	18.4687	18.6843
0.4	16.9272	17.1241	17.3269	17.5297	17.7350
0.5	16.0336	16.2151	16.4011	16.5875	16.7786
0.6	15.1462	15.3095	15.4752	15,6422	15.8151
0.7	14.2652	14.4072	14.5491	14.6938	14.8445
0.8	13.3905	13.5081	13.6229	13.7423	13.8667
0.9	12.5221	12.6124	12.6965	12.7877	12.8819
Section B : x_{Sn}/x_{Bi} = 0.5					
0.1	20.2931	20.4983	20.6959	20.9149	21.1020
0.2	19.3261	19.5268	19.7218	19.9341	20.1235
0.3	18.3614	18.5547	18.7435	18.9466	19.1340
0.4	17.2726	17.4598	17.6495	17.8455	18.0342
0.5	16.4369	16.6067	16.7730	16.9491	17.1205
0.6	15.4779	15.6316	15.7815	15.9399	16.0972
0.7	14.5205	14.6551	14.7852	14.9231	15.0622
0.8	13.5649	13.6775	13.7844	13.8991	14.0159
0.9	12.6117	12.6995	12.7796	12.8684	12.9588
Section C : x_{Sn}/x_{Bi} = 2					
0.1	19.0640	19.2434	19.4227	19.6136	19.7932
0.2	18.1923	18.3773	18.5647	18.7595	18.9495
0.3	17.3323	17.5182	17.7082	17.9022	18.0967
0.4	16.4974	16.7363	16.9754	17.2248	17.4789
0.5	15.6504	15.8238	16.0020	16.1808	16.3664
0.6	14.8274	14.9875	15.1514	15.3156	15.4880
0.7	14.0172	14.1594	14.3032	14.4483	14.6014
0.8	13.2193	13.3388	13.4570	13.5783	13.7063
0.9	12.4332	12.5253	12.6122	12.7052	12.8022

As can be seen in Figure 10 below, the calculated molar volumes Vm are quite similar to those represented by dashed line which indicates additive molar volumes. It can be concluded that molar volume varies with compositions of Ag in a close to linear manner which is characteristic to ideal solution with excess molar volumes are close to zero (Vexc ≈ 0).

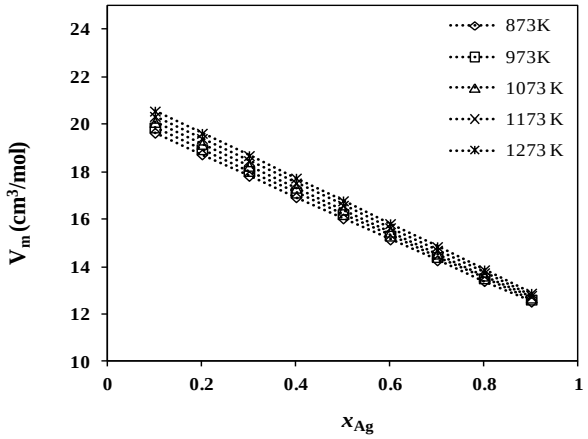

Figure 8: Calculated molar volume isotherms versus molar fraction of Ag for x_{Sn}/x_{Bi}=1/1

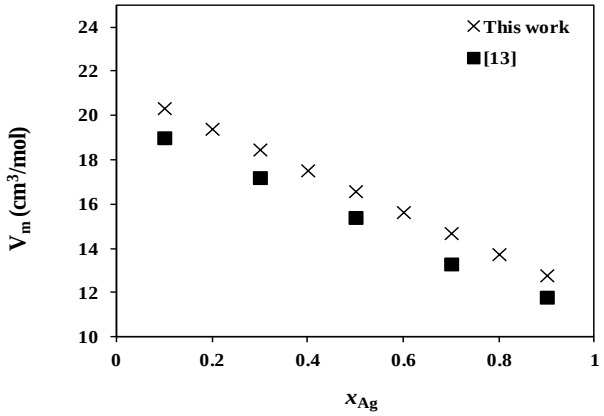

Figure 9: Calculated molar volume versus molar fraction of Ag for x_{Sn}/x_{Bi}=1/1 at 1173 K. Comparison with Fima's values [13]

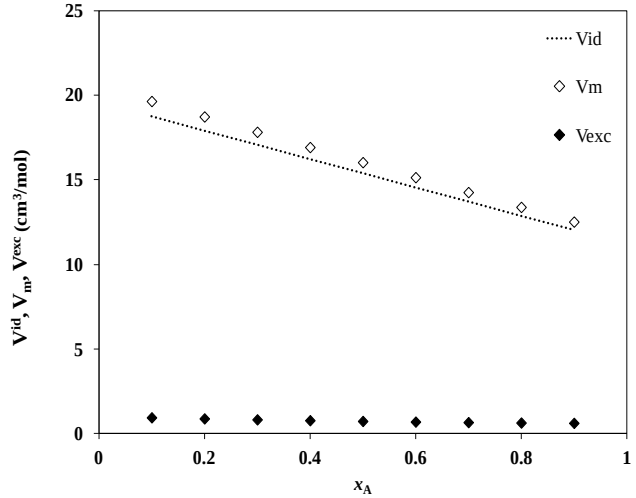

Figure 10: Calculated molar volume V_m versus molar fraction of Ag for x_{Sn}/x_{Bi}=1/1 at 1173 K, together with an excess molar volume (V_{exc}) and an ideal molar volume (V_{id})

Iso-lines for the three physical properties in the ternary system Ag-Bi-Sn at 873 K predicted by Kohler's model [22] for surface tensions and molar volumes, and by Seetharaman and Sichen [28] model for viscosities were calculated as shown in Figures 11-13. It can be noted that the high values of the three investigated physical properties are obtained near the Ag-rich side.

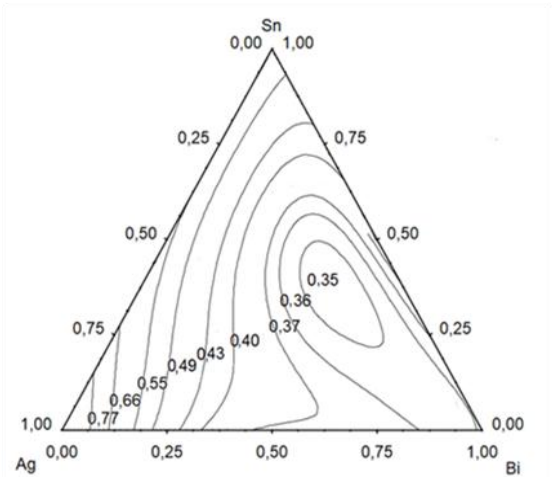

Figure 11: Iso-surface tension curves (N/m) in the liquid Ag-Bi-Sn alloys at 873 K

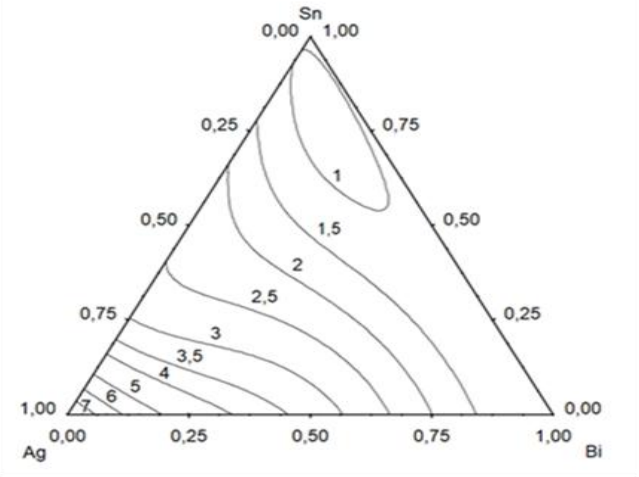

Figure 12: Iso-viscosity curves (mPa.s) in the liquid Ag-Bi-Sn alloys at 873 K

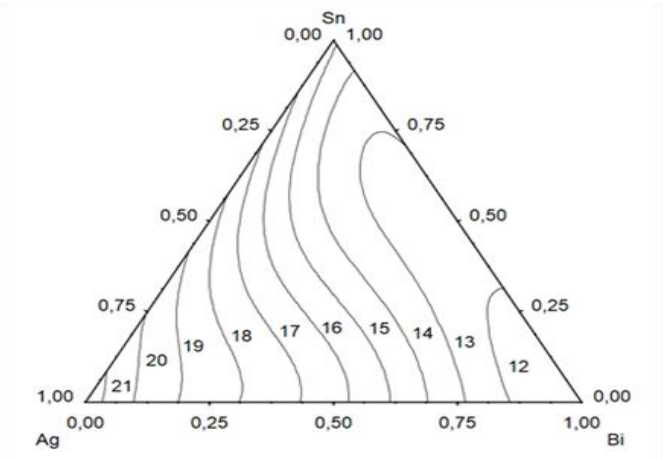

Figure 13: Iso-molar volume curves (cm^3/mol) in the liquid Ag-Bi-Sn alloys at 873 K

Conclusions

In this work, the surface tensions, viscosities and molar volumes of liquid Ag-Bi-Sn alloys were calculated using the Kohler and Seetharaman models. The calculations were done at different temperatures by adding silver to three different sections with x_{Sn}/x_{Bi} = 1/2, 1/1 and 2/1. The results show that up to 40 at. % Ag, no influence of the surface tension and no significant dependence on temperature has been noted. With increasing

temperature the viscosity decreases but increases with Ag-compositions. The molar volumes decrease with increasing Ag-compositions and increase with increasing temperature. It was found that the linear variation of molar volume with Ag-content is characteristic to ideal solution.
In order to confirm the calculated results, it would be helpful to have further experimental data, especially for the viscosity of the Ag-Bi-Sn system where no experimental measurements is available.

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