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Physicochemical Properties of Sb-Sn-Zn Alloys

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In this work, liquid Sb-Sn-Zn alloys were studied to determine their density, viscosity, and surface tension using the discharge crucible method. The measurements were carried out for alloy compositions having X_{Sn}/X_{Sb} ratio of 1, 3, 4, and 9 and Zn content X_{Zn} of 0.05, 0.1, 0.2, and 0.7 in the temperature range from 550 K to 1050 K. The effect of the Zn concentration in the Sb-Sn-Zn alloys on their density, viscosity, and surface tension was observed. Over a wide temperature range, the viscosity and surface tension increased with increasing Zn content in the alloy, while the density decreased. The experimental results obtained for surface tension and viscosity were compared with the results of the Butler model for surface tension and with the Moelwyn-Hughes, Sichen–Boygen–Seetharaman, Seetharaman–Sichen, Kozlov–Romanov–Petrov, and Kaptay models for viscosity.

Key words: Discharge crucible method, Sb-Sn-Zn alloys, density, surface tension, viscosity

INTRODUCTION

The introduction of an European Union Directive and the explorative work on lead solder replacements resulted in interest in Sb-Sn-Zn alloys, which are potential replacements for lead solder. The most popular alloys can be divided into two main groups, i.e., Sn-Ag-Cu (SAC) and Sn-Zn, which are used as replacements for low-temperature lead-based solders (Sn-Pb, Sn-Pb-Cd).[1] Further applications of Sb-Sn-Zn alloys are as a sputtered amorphous layer of compact discs (CDs) for storing information[2] and as a solid-state phase in Zn_4Sb_3- and $SnZnSb_2$-modified InSb,[3] resulting in further interest in this system. The main trend of research is to find replacements for lead solder with low- and high-temperature properties similar to those of toxic solders containing Pb and Cd, which are characterized by melting points above 473 K and 623 K, respectively, for use in different types of connections, not only as solder alloys for pressure soldering, but also for installation of optical components in industries such as automotive, aerospace, electronics, etc.[4]

Eutectic Sn-Zn alloys[5–8] form the second most interesting group of metallic materials for use in the electronics industry, the first being the triple eutectic Ag-Sn-Cu (SAC) alloys.[9–11] The most frequently used solder modifiers for Sn-Zn eutectic are bismuth, indium, and antimony.[12–14] Antimony addition is used to improve the mechanical properties and increase the melting temperature range from 472 K to 500 K.[15] Addition of antimony to SAC with composition Sn-3.5Ag-0.7Cu (at.%) increases its tensile strength,[16] with the best results observed for 1 at.% Sb content. Addition of Sb to eutectic Sn-3.5Ag alloy increases the melting point of the ternary Sn-Ag-Sb alloy[17] and changes the structure of the starting Sn-Ag alloy, with increasing hardness for increasing antimony concentration. In wetting tests, higher Sb content in Sb-Sn alloys resulted in decreases of intermetallic layers.[18] Increases of the melting point and decreases of the density and surface tension with increasing Sb

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content have been observed in ternary Sn-Ag-Sb alloys.[19] Addition of antimony was also observed to improve the mechanical properties of Sn-Ag alloy, with 1.5 at.% Sb content being particularly suitable. According to Miric's[20] study, the poor wettability of Sn-25Ag-10Sb (at.%) alloy on Cu substrate corresponds to the high content of antimony, in contrast to the data obtained in Ref. 19 for Sn-Ag-Sb with Sb content from 8 to 20, for which the contact angle on Cu was almost the same (~33°).

The lack of experimental data on the physicochemical properties of Sb-Sn-Zn alloys prevents their potential use in a wide range of applications. The only data for this system is the projection of the liquidus surface,[21] which was prepared on the basis of data from binary systems without taking into account the impact of ternary data, which were unavailable at that time (1976). The thermodynamic equilibrium relations for liquid Sb-Sn-Zn alloys were described in Ref. 22.

The aim of this work is to analyze new experimental data for the density, surface tension, and viscosity of liquid Sb-Sn-Zn alloys in view of different models.

EXPERIMENTAL PROCEDURES

Sample Preparation

The alloys were prepared from pure Sb, Sn, and Zn (all of purity 99.999%) according to the nominal compositions by the Institute of Electronic Materials Technology and the Experimental Department at Skawina Ironworks. The samples were prepared

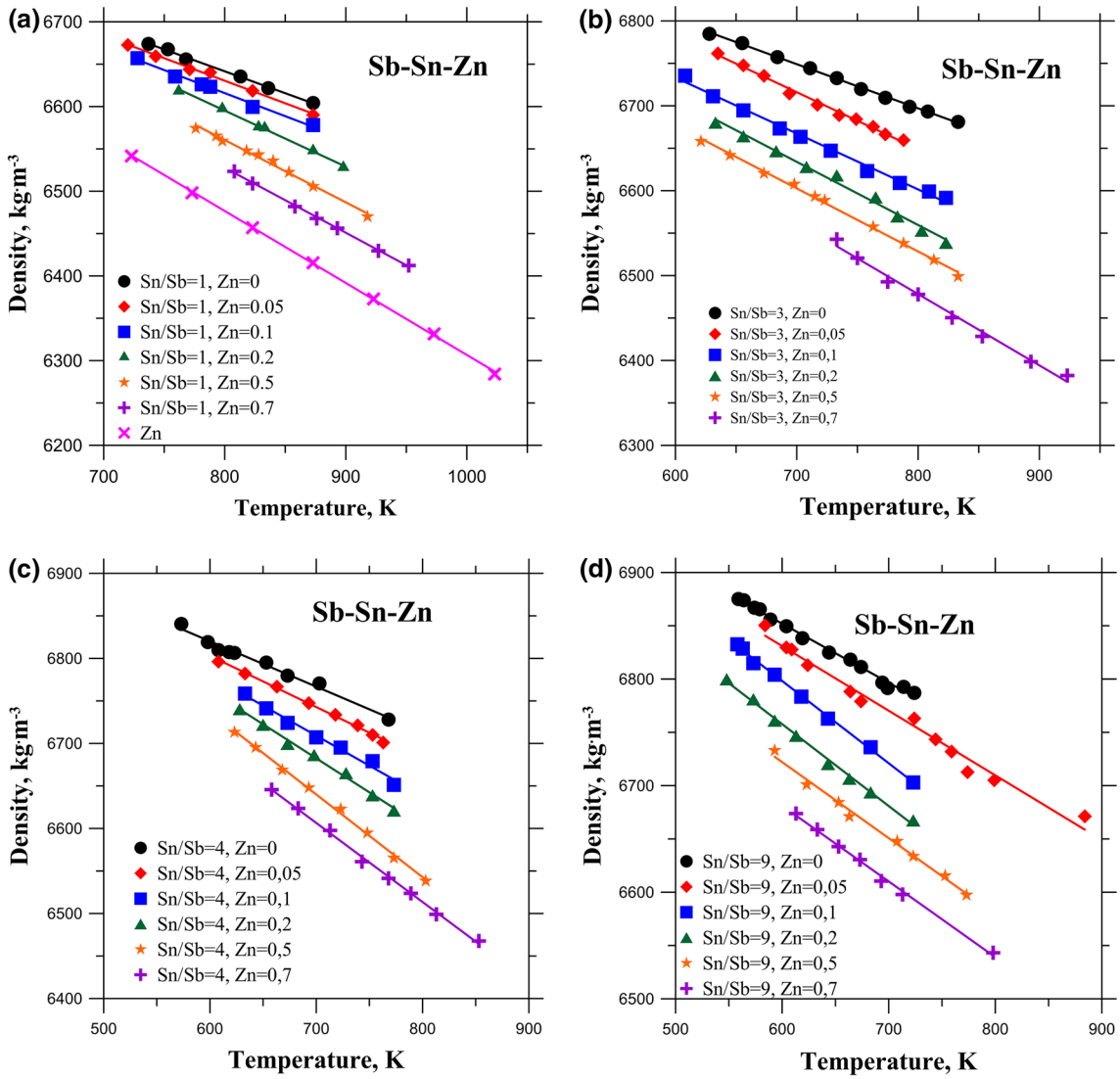

Fig. 1. Density of Sb-Sn-Zn alloys for constant X_{Sn}/X_{Sb} ratio of (a) 1, (b) 3, (c) 4, and (d) 9 with Zn addition.

using an electronic balance to accuracy of 0.1 g with total weight of approximately 1.5 kg. Then, samples were melted in a graphite measuring crucible. The experiment began when the temperature stabilized. For measurements of surface tension, density, and viscosity, the discharge crucible (DC) method was used.[23,24] The study was conducted in a glovebox under a protective atmosphere of 99.9999% pure argon, where the volume concentration of O_2 and H_2O was less than 1 ppm. The atmosphere provided by this equipment ensures that the tests were carried out under stable and repeatable conditions. This method allows one to obtain these three properties from one test at a given temperature. The molar volume was calculated from the experimental density. For the isotherms at 923 K and 1123 K, the presented data were calculated from the measured values.

RESULTS AND DISCUSSION

Figure 1 shows the density of the Sb-Sn-Zn alloys for constant X_{Sn}/X_{Sb} ratio of 1, 3, 4, and 9 with addition of 0.05, 0.1, 0.2, 0.5, 0.7 mole fraction of Zn. The presented data for the densities of pure Zn and the Sb-Sn alloys are taken from Ref. 24. As can be observed from this figure, the densities of the ternary alloys are between the density values of the Sb-Sn alloys and pure Zn.[24] We observed the same trend for all the X_{Sn}/X_{Sb} ratios: with increasing Zn addition in the ternary Sb-Sn-Zn alloys, the density decreased. As can be seen from the isotherm of density at temperature of 923 K (Fig. 2), the composition dependence of the density is not linear, as in the case of the activity of Zn in Sb-Sn-Zn alloys.[23] The density data for the binary alloys are taken from Ref. 25 for Sb-Zn, Ref. 24 for Sb-Sn, and Ref. 26 for Sn-Zn. The molar volumes V_m ($m^3\ mol^{-1}$) of the Sb-Sn-Zn system at 923 K are presented in Fig. 3. The calculations of V_m were carried out using the same equation as in Ref. 24. The observed ternary alloys showed positive deviation from the ideal molar volume. A similar effect was observed for the activity of Zn ternary alloys:[22] over the whole range, there were positive deviations. However, for the alloys with X_{Sn}/X_{Sb} ratio of 1, the variations were very small and increased with increasing Sn content in the alloy.

Figure 4 presents the surface tension of the Sb-Sn-Zn alloys for constant X_{Sn}/X_{Sb} ratio of 1, 3, 4, and 9 with Zn addition. The values of the surface tension are between the values for the Sn-Sb alloys and pure Zn.[24] With increasing Zn content in the ternary Sb-Sn-Zn alloys, the surface tension increased. Figure 5 illustrates the isotherm of the surface tension at temperature of 923 K; it can be seen that the surface tension exhibits a nonlinear

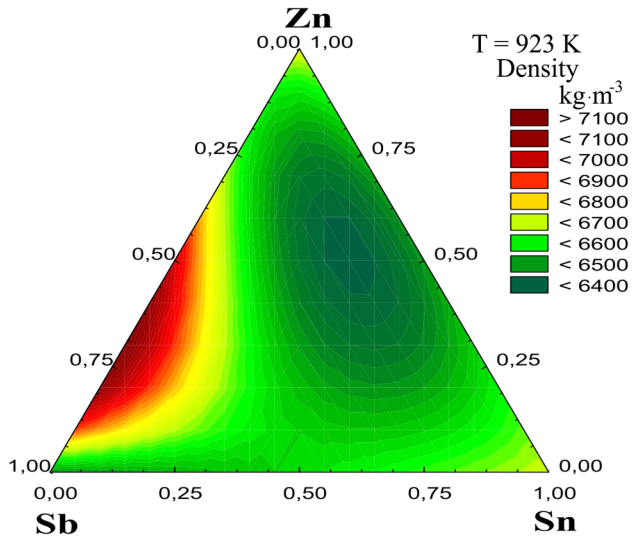

Fig. 2. Isotherm of density of Sb-Sn-Zn alloys at temperature of 923 K.

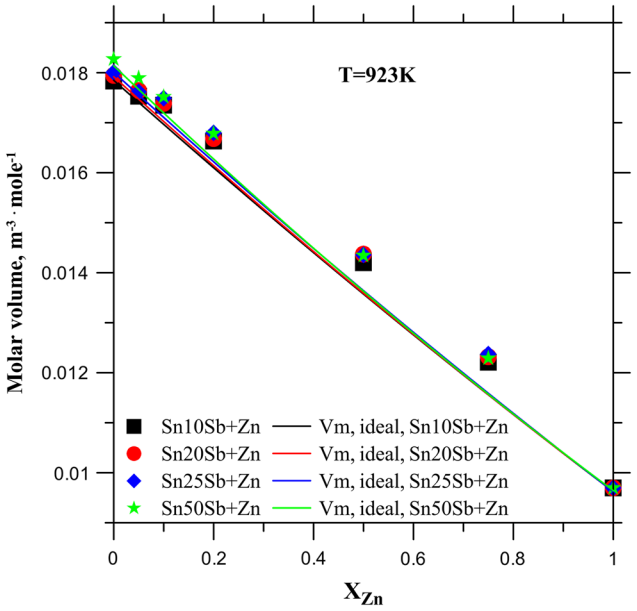

Fig. 3. Molar volumes of the Sb-Sn-Zn system at 923 K.

dependence on the concentration, as for the density. For the prepared isotherm, the data for the binary systems were taken from Ref. 24 for Sb-Sn, Ref. 27 for Sb-Zn, and Ref. 26 for Sn-Zn.

Linear equations for the temperature dependence of the density (Table I) and surface tension (Table II) were determined using the least-squares method, and the coefficients A and B and the errors of these coefficients were calculated. In addition, values were calculated for temperature of 923 K. An

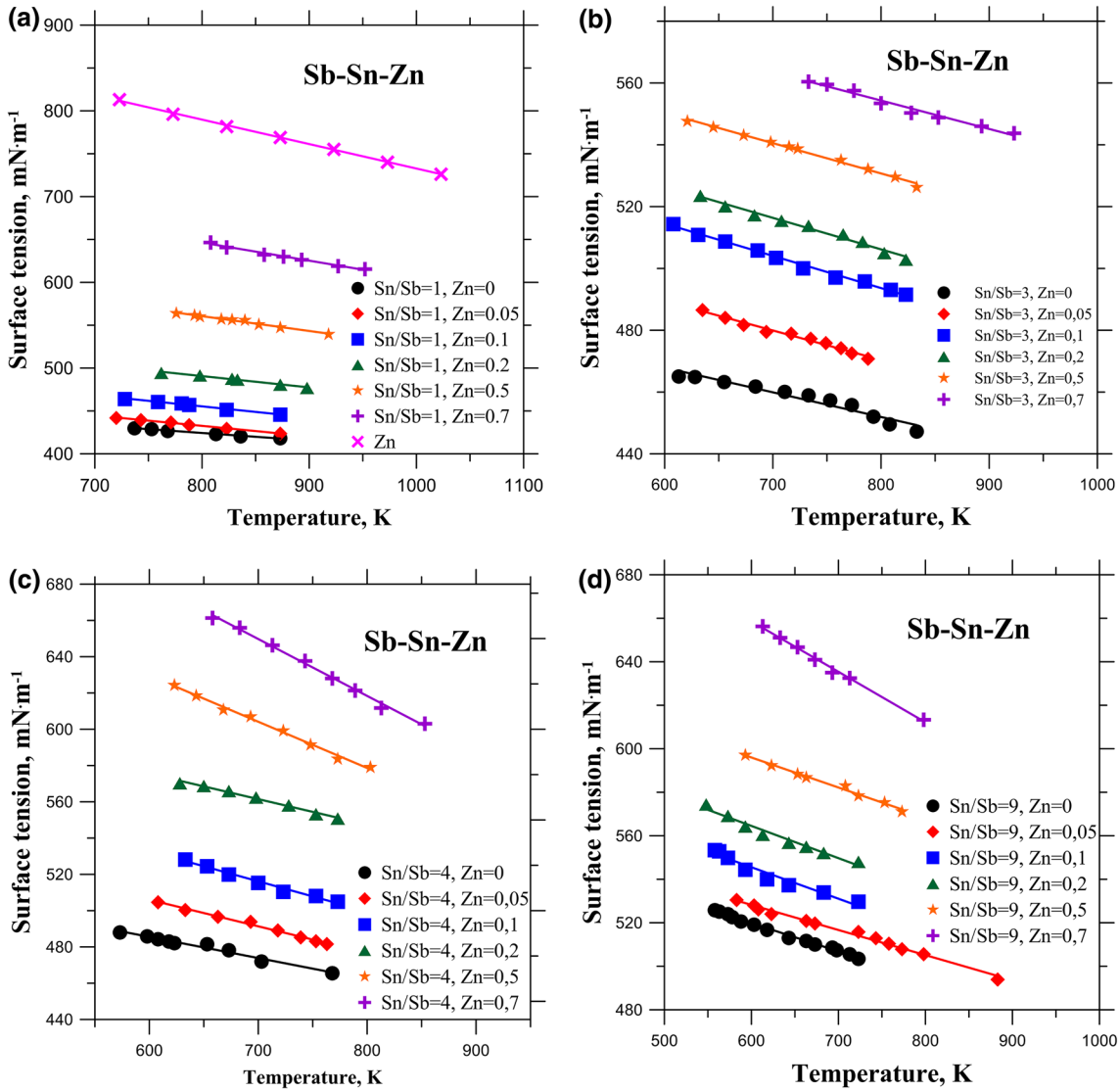

Fig. 4. Surface tension of Sb-Sn-Zn alloys for constant X_{Sn}/X_{Sb} ratio of (a) 1, (b) 3, (c) 4, and (d) 9 with Zn addition.

Arrhenius equation describes the viscosity with a designated coefficient A and activation energy E. The viscosity values calculated for temperature of 923 K are summarized in Table III.

The experimentally obtained values of viscosity for the Sb-Sn-Zn alloys are shown in Fig. 6. The viscosity values obtained for the ternary alloys are between the values for pure Zn and the binary Sb-Sn alloys.[24] Figure 7 shows the isotherm of viscosity at temperature of 923 K, and Fig. 8 presents a three-dimensional (3D) projection of the viscosity at temperature of 1123 K, which better shows the minima and maxima of the viscosity values of the Sb-Sn-Zn system. These areas are associated with short-range ordering of the ternary $SnZnSb_2$ phase. The viscosity data for the binary alloys were taken from Refs. 28 and 29 for Sb-Zn, Ref. 24 for Sb-Sn, and Refs. 30 and 31 for Sn-Zn.

The results of the experiments were compared with Butler's[32] model (for surface tension), and with the Moelwyn-Hughes,[33] Sichen–Boygen–Seetharaman,[34] Seetharaman–Sichen,[35] Kozlov–Romanov–Petrov,[36] and Kaptay[37] models (for viscosity) for the two temperatures (923 K and 1123 K).

The Butler model,[32] whose theoretical foundations are discussed in Ref. 23, was used for modeling the surface tension. Figures 9, 10, 11, and 12 show the values calculated using the Butler model together with the experimental data for the surface tension for the two temperatures of 923 K and 1123 K for X_{Sn}/X_{Sb} ratio of 9, 4, 3, and 1 with Zn addition. For the modeling of the surface tension, the density and surface tension of the pure metals determined experimentally in Ref. 24 were used. The optimized thermodynamic parameters for the binary systems used in the model were derived from the European Cooperation in Science and Technology COST 531 database.[38] There was good agreement between the values obtained from the Butler model and the experimental surface tension data for all the alloys with given X_{Sn}/X_{Sb} ratio, although the compatibility seemed to be better at temperature of 923 K than 1123 K. For the alloys with X_{Sn}/X_{Sb} ratio of 1 at temperature of 923 K and 1123 K, the effect was almost identical.

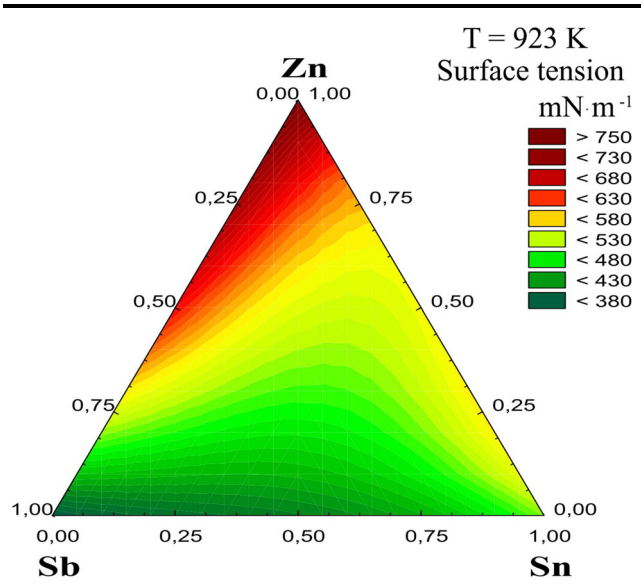

Fig. 5. Isotherm of surface tension of Sb-Sn-Zn alloys at temperature of 923 K.

Isotherms for viscosity were calculated for the Sb-Sn-Zn alloys using the Moelwyn-Hughes,[33] Sichen–Boygen–Seetharaman,[34] Seetharaman–Sichen,[35] Kozlov–Romanov–Petrov,[36] and Kaptay[37] models and are presented in Figs. 13, 14, 15, and 16. The model was discussed in Ref. 23. The calculation results were compared with their experimental data. Calculations were performed for the Sb-Sn-Zn alloys at constant X_{Sn}/X_{Sb} ratio of 9, 4, 3, and 1 and variable Zn content at temperatures of 923 K and 1123 K. The values of the viscosity, density, and surface tension of the pure metals used in the models were determined experimentally in Ref. 24. The thermodynamic properties used in the model come from the COST 531 database.[38] None of the proposed models describes the viscosity values obtained in the experiment in an optimum way. The best compliance between the experimental viscosity data and the modeled values was obtained from

Table I. Density of Sb-Sn-Zn alloys

Chemical Composition			$\rho = A + BT$ (kg m^{-3})					
X_{Sb}	X_{Sn}	X_{Zn}	A	δA	B	δB	ρ (923 K)	$\delta\rho$
0.095	0.855	0.05	7194.9	58.2	−0.606	0.014	6635.6	58.2
0.09	0.81	0.1	7261.6	5.6	−0.772	0.009	6549.0	33.4
0.08	0.72	0.2	7218.7	10.2	−0.769	0.011	6508.9	60.4
0.05	0.45	0.5	7150.3	17.1	−0.714	0.011	6491.3	112.5
0.03	0.27	0.7	7107.6	10.8	−0.711	0.008	6451.3	43.9
0.19	0.76	0.05	7164.5	4.9	−0.602	0.009	6608.9	29.6
0.18	0.72	0.1	7203.5	25.1	−0.706	0.029	6551.9	125.4
0.16	0.64	0.2	7242.9	12.3	−0.801	0.014	6503.6	61.4
0.1	0.4	0.5	7321.2	8.9	−0.973	0.011	6423.1	53.4
0.06	0.24	0.7	7257.5	10.9	−0.930	0.012	6399.1	43.9
0.2375	0.7125	0.05	7186.4	9.4	−0.606	0.013	6627.0	65.3
0.225	0.65	0.1	7128.5	20.2	−0.659	0.021	6520.2	74.9
0.2	0.6	0.2	7154.7	25.2	−0.744	0.024	6467.9	161.7
0.125	0.375	0.5	7122.6	9.1	−0.743	0.011	6436.8	176.8
0.075	0.225	0.7	7150.3	35.6	−0.840	0.026	6374.9	72.1
0.475	0.475	0.05	7056.3	19.8	−0.532	0.016	6565.3	22.9
0.45	0.45	0.1	7051.4	19.1	−0.544	0.019	6549.3	29.9
0.4	0.4	0.2	7124.5	13.9	−0.666	0.015	6509.6	13.4
0.25	0.25	0.5	7148.0	21.4	−0.735	0.011	6469.6	71.8
0.15	0.15	0.7	7141.5	11.1	−0.767	0.01	6433.6	10.3

Table II. Surface tension of Sb-Sn-Zn alloys

Chemical Composition			$\sigma = A + BT$ (mN m^{-1})					
X_{Sb}	X_{Sn}	X_{Zn}	A	δA	B	δB	σ (923 K)	$\delta\sigma$
0.095	0.855	0.05	597.5	1.2	−0.115	0.001	491.4	11.5
0.09	0.81	0.1	632.4	4.8	−0.145	0.006	498.6	18.9
0.08	0.72	0.2	653.4	2.6	−0.148	0.004	516.8	15.9
0.05	0.45	0.5	678.7	1.0	−0.138	0.001	551.3	5.8
0.03	0.27	0.7	797.8	1.1	−0.232	0.001	583.7	5.6
0.19	0.76	0.05	593.9	0.6	−0.146	0.001	459.1	3.7
0.18	0.72	0.1	632.5	1.2	−0.166	0.003	479.3	7.4
0.16	0.64	0.2	659.8	0.8	−0.140	0.001	530.6	3.8
0.1	0.4	0.5	782.6	1.2	−0.255	0.002	547.2	9.7
0.06	0.24	0.7	869.3	2.4	−0.313	0.005	580.4	14.3
0.2375	0.7125	0.05	546.7	3.9	−0.095	0.005	459.01	3.8
0.225	0.65	0.1	577	3.5	−0.104	0.003	481.00	3.5
0.2	0.6	0.2	587.8	5.9	−0.102	0.008	493.65	8.9
0.125	0.375	0.5	609.2	2.9	−0.098	0.002	518.74	2.9
0.075	0.225	0.7	627.9	4.9	−0.092	0.007	542.98	4.9
0.475	0.475	0.05	529.9	0.9	−0.121	0.001	418.2	1.9
0.45	0.45	0.1	558.8	2.6	−0.129	0.003	439.7	2.6
0.4	0.4	0.2	596.3	2.8	−0.132	0.004	474.5	2.8
0.25	0.25	0.5	698.4	8.0	−0.172	0.009	539.6	8.0
0.15	0.15	0.7	814.9	1.9	−0.211	0.002	620.1	9.7

Table III. Viscosity of Sb-Sn-Zn alloys

Chemical Composition			$\eta = A'e^{E/RT}$ (mPa s)					
X_{Sb}	X_{Sn}	X_{Zn}	A	δA	E	δE	η (923 K)	$\delta\eta$
0.095	0.855	0.05	0.329	0.010	9304.2	50.2	1.106	0.011
0.09	0.81	0.1	0.245	0.009	11,482.5	73.1	1.094	0.009
0.08	0.72	0.2	0.261	0.005	11,656.2	49.3	1.192	0.032
0.05	0.45	0.5	0.339	0.007	11,237.2	22.9	1.466	0.040
0.03	0.27	0.7	0.326	0.006	12,306.4	48.7	1.621	0.018
0.19	0.76	0.05	0.248	0.005	11,086.7	30.7	1.052	0.015
0.18	0.72	0.1	0.301	0.003	10,552.1	22.8	1.191	0.019
0.16	0.64	0.2	0.321	0.001	10,633.6	19.3	1.283	0.017
0.1	0.4	0.5	0.298	0.039	11,646.3	146.7	1.359	0.069
0.06	0.24	0.7	0.292	0.029	12,747.9	105.4	1.538	0.052
0.2375	0.7125	0.05	0.305	0.001	9901.2	16.4	1.108	0.016
0.225	0.65	0.1	0.433	0.019	8395.5	48.9	1.293	0.019
0.2	0.6	0.2	0.549	0.018	7650.3	44.8	1.487	0.041
0.125	0.375	0.5	0.696	0.031	6840.8	65.1	1.697	0.038
0.075	0.225	0.7	0.890	0.008	5947.1	26.7	1.931	0.024
0.475	0.475	0.05	0.292	0.001	11,052.9	19.6	1.233	0.013
0.45	0.45	0.1	0.273	0.003	11,924.7	55.2	1.291	0.017
0.4	0.4	0.2	0.319	0.002	11,310.9	40.7	1.393	0.012
0.25	0.25	0.5	0.293	0.001	12,757.0	20.1	1.545	0.015
0.15	0.15	0.7	0.38	0.002	12,088.8	46.5	1.836	0.018

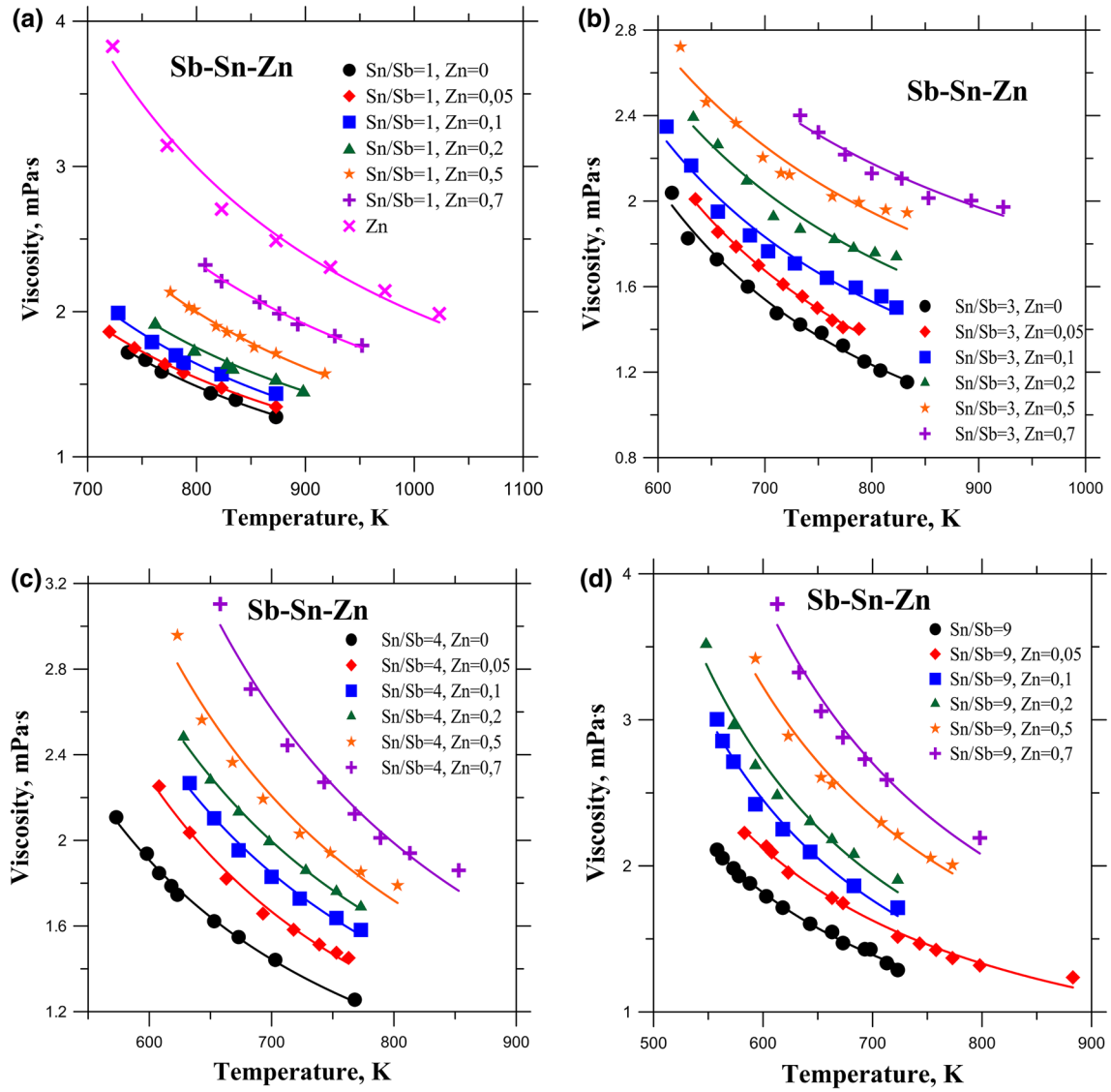

Fig. 6. Viscosity of Sb-Sn-Zn alloys for constant X_{Sn}/X_{Sb} ratio of (a) 1, (b) 3, (c) 4, and (d) 9 with Zn addition.

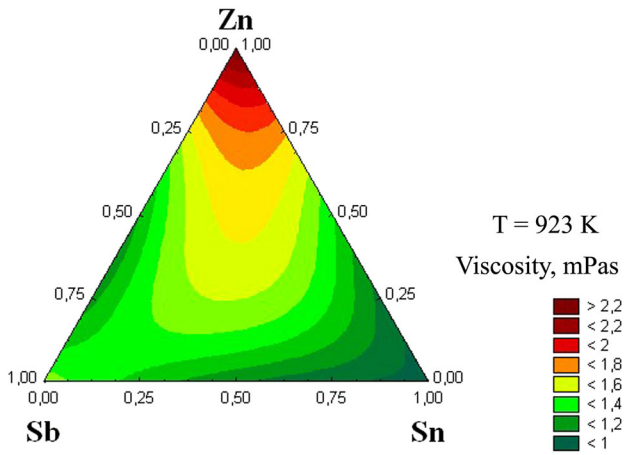

Fig. 7. Isotherm of viscosity of Sb-Sn-Zn alloys at temperature of 923 K.

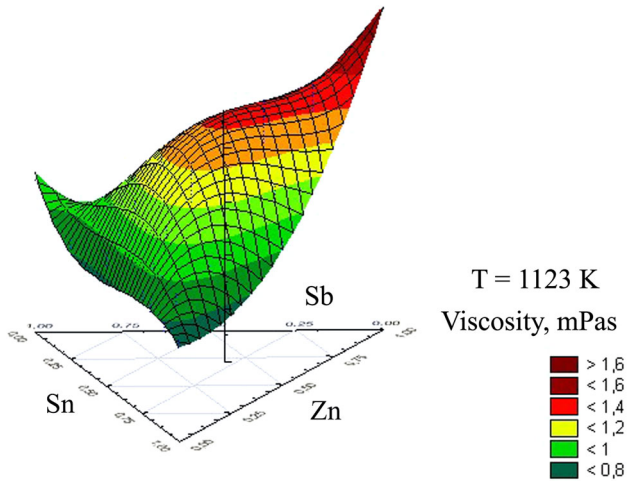

Fig. 8. Three-dimensional projection of viscosity of Sb-Sn-Zn alloys at temperature of 1123 K.

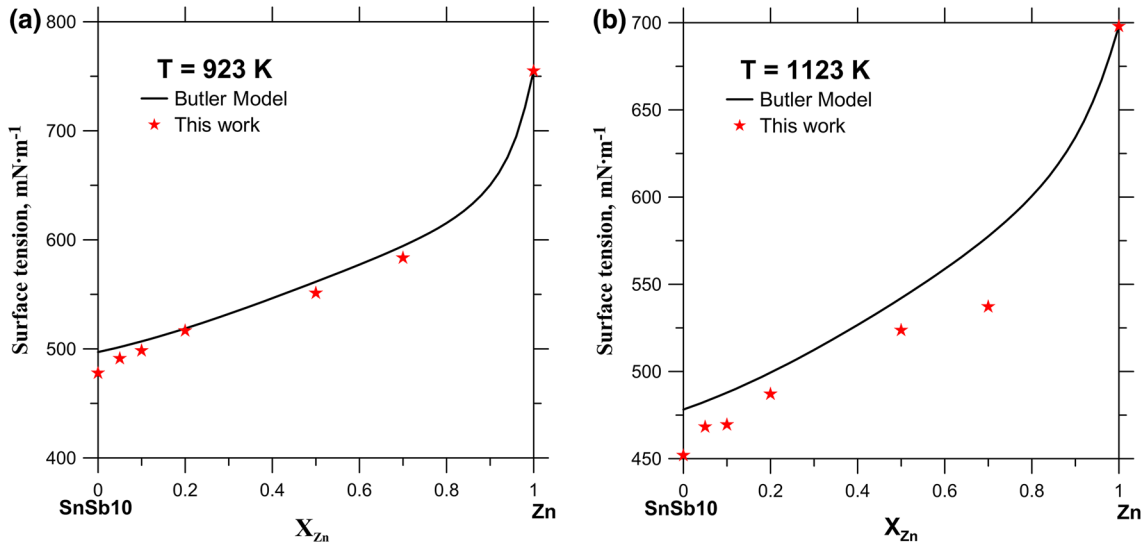

Fig. 9. Experimental surface tension data compared with the Butler model for Sb-Sn-Zn alloys with X_{Sn}/X_{Sb} ratio of 9 at temperature of (a) 923 K and (b) 1123 K.

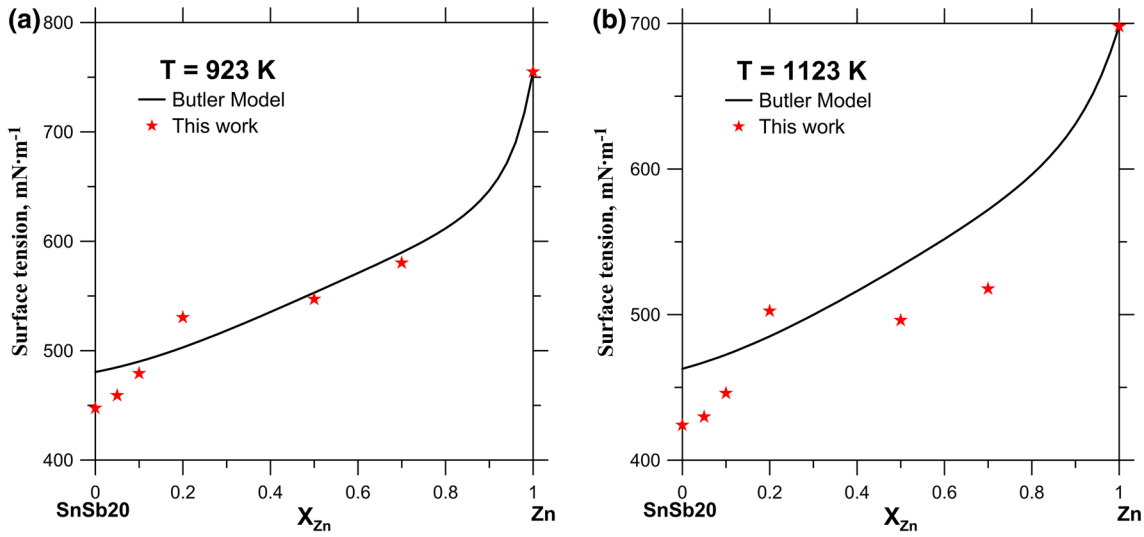

Fig. 10. Experimental surface tension data compared with the Butler model for Sb-Sn-Zn alloys with X_{Sn}/X_{Sb} ratio of 4 at temperature of (a) 923 K and (b) 1123 K.

three models. The first and second are the Kaptay[37] and Kozlov–Romanov–Petrov[36] models for X_{Sn}/X_{Sb} ratio of 1 at 923 K and 1123 K. The third is the Sichen–Boygen–Seetharaman[34] model for X_{Sn}/X_{Sb} ratio of 9 at 1123 K and for X_{Sn}/X_{Sb} ratio of 4 at 923 K and 1123 K.

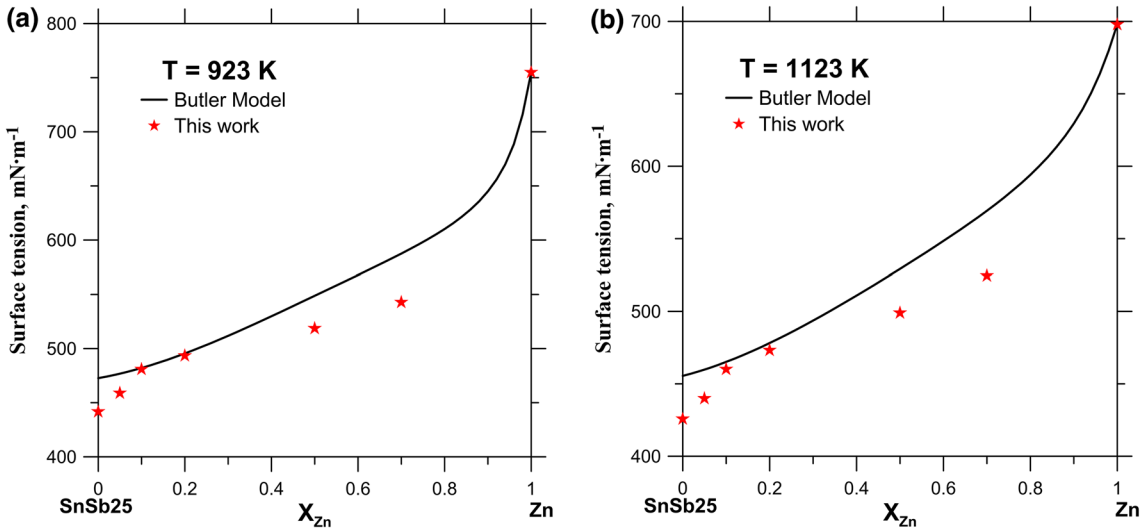

Fig. 11. Experimental surface tension data compared with the Butler model for Sb-Sn-Zn alloys with X_{Sn}/X_{Sb} ratio of 3 at temperature of (a) 923 K and (b) 1123 K.

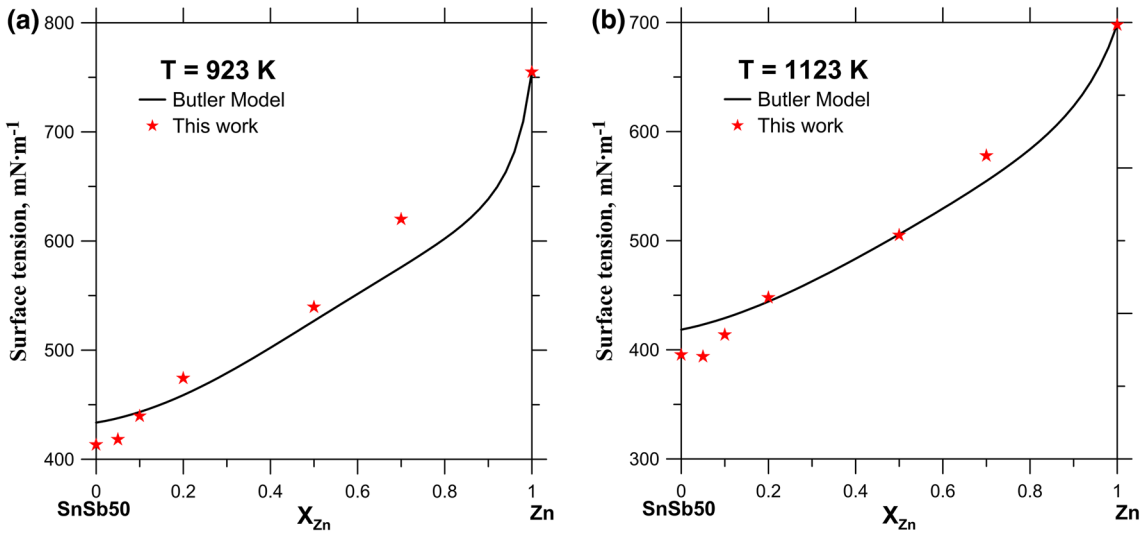

Fig. 12. Experimental surface tension data compared with the Butler model for Sb-Sn-Zn alloys with X_{Sn}/X_{Sb} ratio of 1 at temperature of (a) 923 K and (b) 1123 K.

From the isotherms for viscosity at the temperatures of 923 K (Fig. 16a) and 1123 K (Fig. 16b) for constant X_{Sn}/X_{Sb} ratio of 1, one can see that the trend of the viscosity is close to linear with Zn addition in the Sb-Sn-Zn alloys. However, for fixed X_{Sn}/X_{Sb} ratio of 9, 3, and 4, one sees a different trend

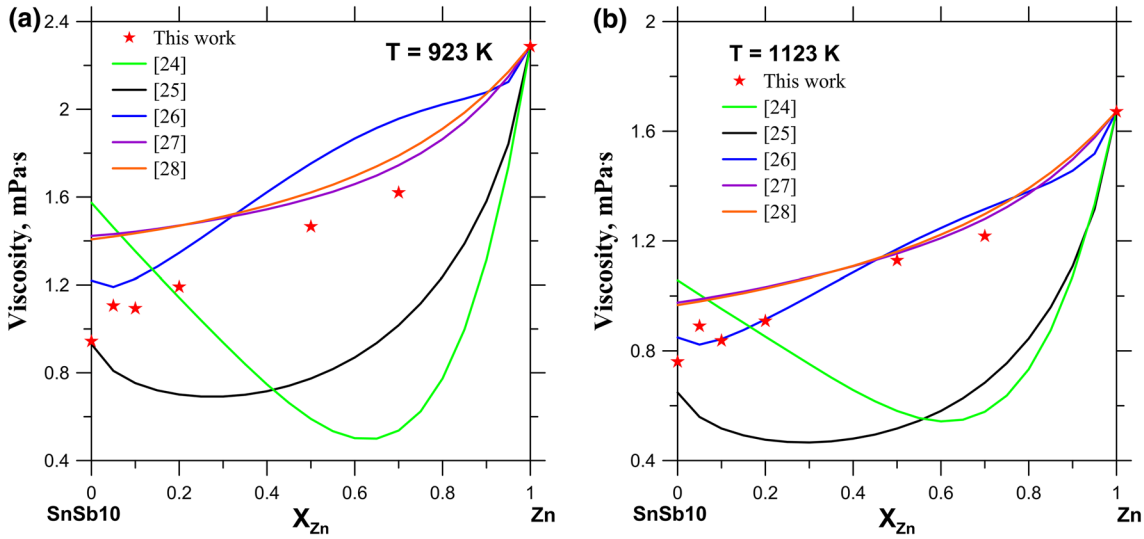

Fig. 13. Experimental viscosity data compared with the viscosity models for Sb-Sn-Zn alloys with X_{Sn}/X_{Sb} ratio of 9 at temperature of (a) 923 K and (b) 1123 K.

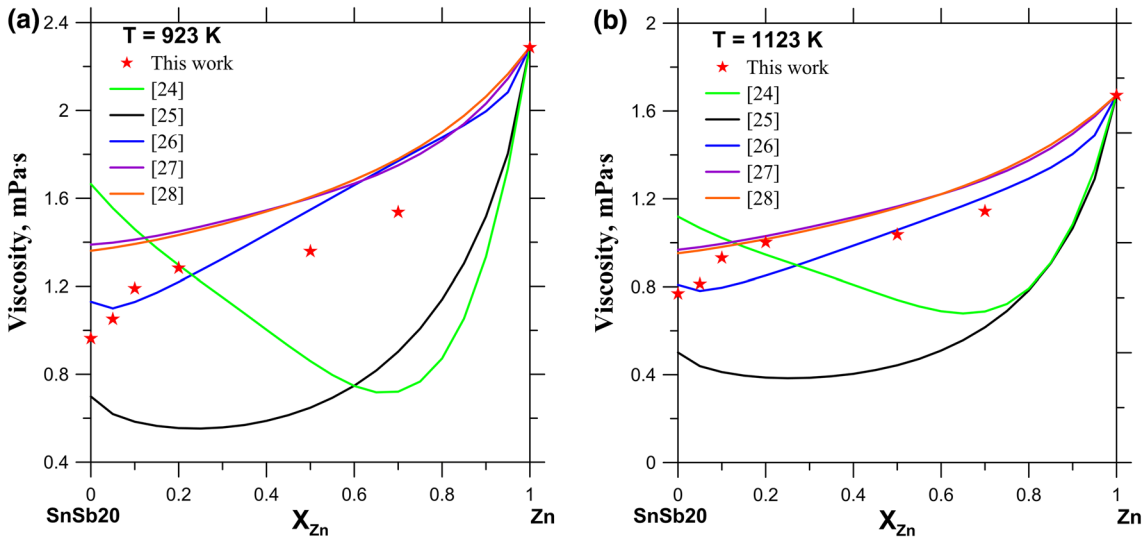

Fig. 14. Experimental viscosity data compared with the viscosity models for Sb-Sn-Zn alloys with X_{Sn}/X_{Sb} ratio of 4 at temperature of (a) 923 K and (b) 1123 K.

in the viscosity. Such trends in viscosity values were also described by Sinha and Miller[28] and Yao,[29] who studied the Sb-Zn system. Those authors[28,29] explained the observed concentration dependence of the viscosity based on short-range ordering in the liquid (maybe associated with the presence of asso-

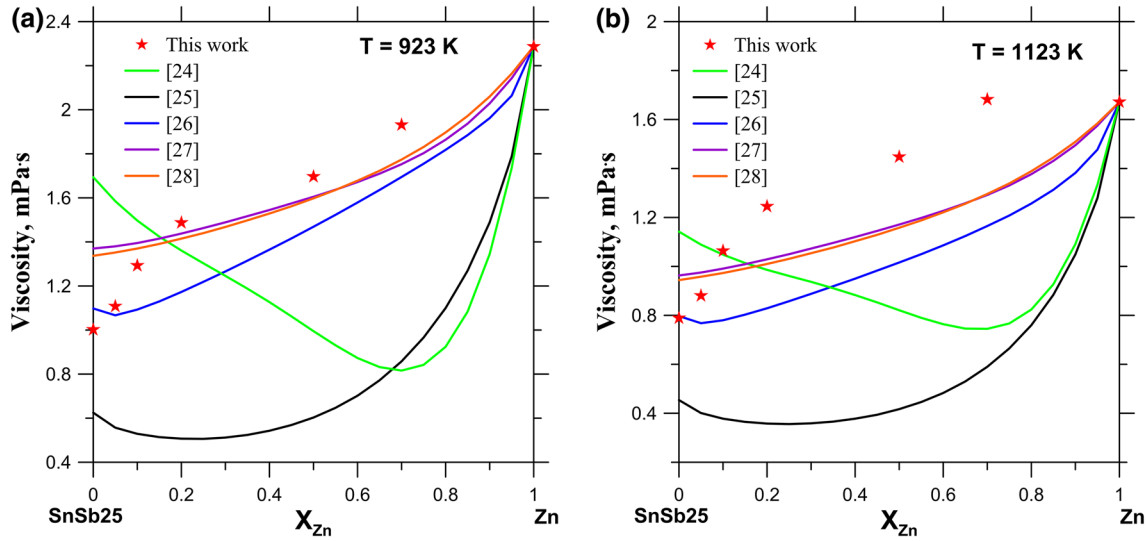

Fig. 15. Experimental viscosity data compared with the viscosity models for Sb-Sn-Zn alloys with X_{Sn}/X_{Sb} ratio of 3 at temperature of (a) 923 K and (b) 1123 K.

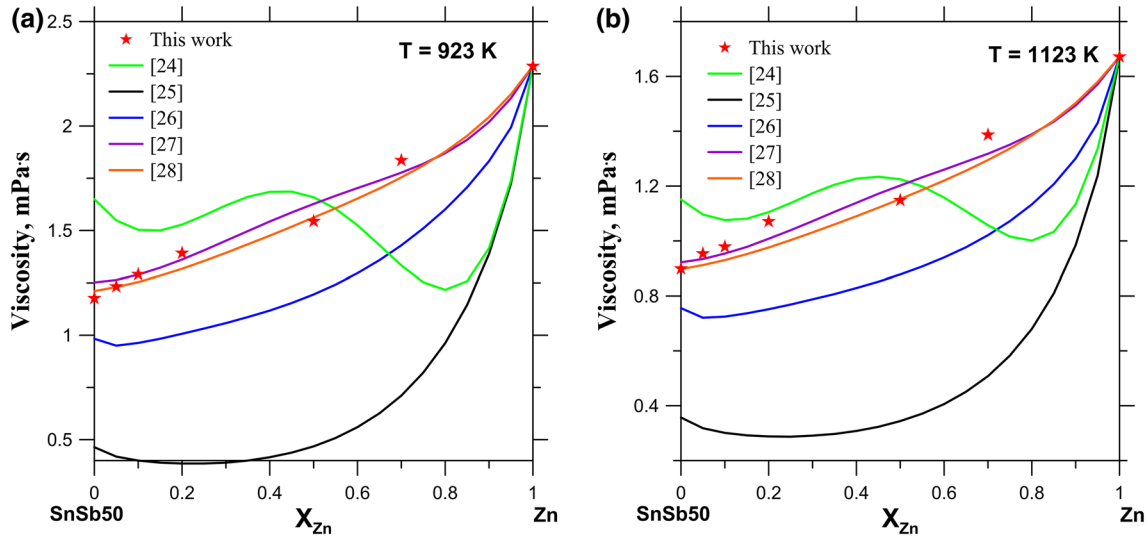

Fig. 16. Experimental viscosity data compared with the viscosity models for Sb-Sn-Zn alloys with X_{Sn}/X_{Sb} ratio of 1 at temperature of (a) 923 K and (b) 1123 K.

ciates with composition similar to the intermetallic composition), resulting in an increase in the value of the viscosity.

CONCLUSIONS

The presented experimental results show that the density of all the tested alloys decreased with increasing temperature and with increasing content of zinc and antimony, whereas the surface tension and viscosity of all the alloys decreased with increasing temperature but increased with increasing zinc content. The measured results for the surface tension show good agreement with the Butler model. For viscosity, the Kaptay[37] and Kozlov–Romanov–Petrov[36] models are in agreement with the experimental measurements.

Such wide knowledge of the physicochemical properties of these alloys could extend the possible applications of this system, and the presented data could be used for designing new lead-free solder materials.

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