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The effects of Na addition on the density, surface tension and viscosity of liquid Sn–Zn alloys

Tomasz Gancarz*, Wladyslaw Gasior

Institute of Metallurgy and Materials Science, Polish Academy of Sciences, Krakow, Poland

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ABSTRACT

In this work, data for liquid SnZn alloys with Na addition are presented. The thermophysical properties, viscosity, density and surface tension were measured using the discharge crucible method (DC). The experiments were conducted for $Sn_{15}Zn$ alloys with 0.1, 0.2, 0.5, 1.0, 3.0 and 5.0 at % Na. The measurements were performed over a temperature range of 548–823 K. The results show that the addition of Na to $Sn_{15}Zn$ (at. %) causes increased density and viscosity, and decreased surface tension in comparison to eutectic SnZn alloy. The new surface tension experimental data were compared with the corresponding values calculated by the Butler model, while the viscosity data were compared with models predicted values obtained by the Sichen-Boygen-Seetharaman, Seetharaman-Sichen, Kozlov-Romanov-Petrov, Kaptay and Gasior models, using thermodynamic input data. Correlation analysis between modelled and experimental data was performed.

1. Introduction

The investigation into lead-free solder alloys, and the development of new alloy systems have been continuing for years in many laboratories all over the world, with the aim of obtaining solders with properties similar to those of traditional Pb–Sn solders [1]. However, producing the solder(s) that possess the many properties comparable to a lead one has long been a great challenge for researchers. Experimental studies on the influence of any metals used instead of lead in solders, on different properties of $(Ag–Sn)_{eut}$ [2], have been also performed in our laboratory [3,4]. These studies aimed to determine the impact of third, fourth and fifth elements in lead-free $(Ag–Sn)_{eut}$ solders.

Many problems linked with silver-tin based solders concern the much higher melting temperature in comparison to the lead containing solders, due to the greater number of defected electronic packages. Regarding melting temperature, the SnZn eutectic based alloys, with a meting point about 20° lower than that of $(Ag–Sn)_{eut}$ and $(Ag–Sn–Cu)_{eut}$ based solders seems to be an attractive substitute for both lead and lead-free solders. Therefore, the Sn–Zn eutectic has been one of the most important groups of alloys under consideration [5–8], so, also at our institute research has been conducted on lowering the melting point, improving wettability [9], and reducing the costs. To designate the best alloys on lead replacement solder, there must be a consistent thermophysical properties database, which allows for the initial designation of alloys fulfilling the necessary criteria.

In recent years, the data of density, viscosity and surface tension for Sn–Zn–Cu [5], SAC [10], SAC + Bi and Sn–Zn [6], Sn–Ag [1,11,12], Sn–Sb [13–15] have been reported. The effects of Na addition on the properties of the Sn–Zn eutectic alloy, such as the melting temperature, linear thermal expansion, electrical resistivity and mechanical properties, together with the microstructural analysis of the cast alloy, were investigated and are presented in Ref. [16].

In the present work, the density, surface tension and viscosity of liquid Sn–Zn alloys containing different amounts of Na were investigated and compared with model predictions, due to a lack of available experimental data.

2. Experimental

Ternary alloys were prepared by melting accurately weighed amounts of high-purity metals (Sn, Zn, 99.999% and Na 99.99%) in a glove-box under a protective atmosphere of high purity argon, with water vapour, nitrogen and oxygen concentration lower than 0.1 ppm to avoid the oxidation of liquid alloys. The $Sn_{15}Zn$ eutectic alloys with 0.1%, 0.2%, 0.5%, 1.0%, 3.0% and 5.0% Na (at. %) addition

* Corresponding author. Tel.: +48 12295284; fax: +48 122952804.
E-mail address: tomasz.gancarz@imim.pl (T. Gancarz).

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were melted in a graphite crucible, and after casting they were used for measuring density, viscosity and surface tension by the discharge crucible method (DC), as described in detail elsewhere [2,17]. The method consists of allowing the free flow of the liquid alloy through a hole in the bottom of the crucible, which is on a set of scales. Based on the measured weight over a given period time, we can calculate the density, surface tension and viscosity. The relation describing the volumetric flow rate of liquid Q, exiting the crucible through the orifice of fixed radius r_0, the head of the liquid is calculated using the equation (1):

$$h = \frac{1}{2g}\left(\frac{Q}{\left(a_4\left(\frac{2\rho Q}{\pi r_0 \eta}\right)^3 + a_3\left(\frac{2\rho Q}{\pi r_0 \eta}\right)^2 + a_2\left(\frac{2\rho Q}{\pi r_0 \eta}\right) + a_1\right)\pi r_0^2}\right) + \frac{\sigma}{\rho g r_0} \quad (1)$$

where: ρ – density of liquid (kg/m^3), g –gravitational acceleration (m/s^2), r_0 – orifice radius in the bottom of crucible (m), σ – surface tension (mN/m), Q – free flow (m^3/s), η and σ viscosity and surface tension, respectively, C_d – discharge coefficient determined for a given crucible, based on the free flow of liquids with known density, Re – Reynolds number and a_1, a_2, a_3 and a_4 are constants in the polynomial describing C_d versus Re.

The cumulative weight of the liquid, C_m, that is the mass of liquid that flowed freely from the crucible in time t, is measured in the course of the experiment, and is commonly described by:

$$C_m = \sum_{i=1}^{n} C_i t^{i-1} \quad (2)$$

and is used for the calculation of free flow Q in accordance with Eq. (3):

$$Q = \frac{C_3 t^2 + C_2 t + C_1}{\rho} \quad (3)$$

where: C_i are the parameters calculated, based on the data C_m and time t.

Using the numerical solutions obtained from equation (1), the density, surface tension and viscosity were calculated for each alloy [2,17]. The crucibles used in these experiments have the same dimensions as those used in our previous work [2,6,13,17].

The temperature dependence of density and surface tension were described by linear equations, and the viscosity by an Arrhenius type equation. The data are listed in Tables 1–3, along with estimated errors of the equation parameters and property values calculated at 573 K, respectively.

Table 1
The temperature dependences of density for $(SnZn)_{eut}$-Na alloys, with the A and B parameters of linear equations describing experimental data and its errors δ and density values calculated at 573 K.

SnZn + X_{Na} (at. %)	A	δA	B	δB	ρ (573 K)	δρ
0.1 Na	7.378	0.007	−0.000772	0.000010	6.936	0.001
0.2 Na	7.375	0.016	−0.000772	0.000025	6.933	0.000
0.5 Na	7.372	0.010	−0.000793	0.000016	6.918	0.001
1.0 Na	7.370	0.010	−0.000830	0.000016	6.894	0.002
3.0 Na	7.364	0.008	−0.000987	0.000011	6.798	0.005
5.0 Na	7.354	0.008	−0.001119	0.000039	6.712	−0.008

Table 2
The temperature dependences of the surface tension for the$(SnZn)_{eut}$-Na alloys with the parameters A and B of linear relationship and their estimated errors δ and surface tension values calculated at 573 K, with value from the Butler model [26] at temperature 573 K.

SnZn + X_{Na} (at. %)	A	δA	B	δB	σ (573 K)	δσ	Butler
0.1 Na	584.0	2.1	−0.070	0.003	543.94	0.14	532.4
0.2 Na	582.6	1.8	−0.076	0.002	539.20	0.50	516.7
0.5 Na	581.2	1.2	−0.086	0.002	532.19	0.51	493.1
1.0 Na	579.2	2.3	−0.098	0.003	523.28	0.62	473.7
3.0 Na	577.9	1.7	−0.116	0.002	511.79	0.19	440.8
5.0 Na	576.8	1.7	−0.132	0.002	501.11	0.19	425.5

3. Results and discussion

3.1. Density

Before the start of experiments, the phase diagram Na–Sn [18], Na–Zn [19], Sn–Zn [20] and DSC measurements data [16] were analysed, and the temperature range for the investigation was established at 548–823 K. The obtained data of surface tension, density and viscosity are shown in Tables 1–3 and in Figs. 1–4. Temperature dependencies of the $(SnZn)_{eut}$-Na alloys density are showed in Fig. 1. As is generally observed in many systems, the density decreases with the rise in temperature, and the increase of Na addition to the SnZn eutectic also influences the density decrease. The isotherm at 573 K, presented in Fig. 2, shows that ternary SnZnNa liquid solutions are characterised by higher density (dashed line) than ideal solutions (solid line). This means that the addition of Na to the SnZn eutectic influences the decrease of the middle interatomic distance between atoms forming alloys. The isotherm of density shown in Fig. 2 at 573 K as a dashed line was calculated using Eq. (4) (shown below) which was worked out using the experimental density obtained in this study and the density of Na, Sn and Zn taken from papers [21] and [6], respectively. All alloys discussed are characterised by lower density in comparison to that for SnPb eutectic (8.18 (g cm^{-3})) [17] at 573 K, and they change at the same temperature from 6.7 to 6.93 (g cm^{-3}) with the increase of Na concentration.

A temperature-concentration dependence of the density of $(SnZn)_{eut}$ liquid alloys with Na additions was generated by the minimisation method, and can be expressed by the following relation:

$$\rho = \rho_{SnZn}(1 - X_{Na}) + \rho_{Na}X_{Na} + \left(0.0139 + 2.4\cdot10^{-5}T\right)X_{Na} + \left(-0.0028 - 4.9\cdot10^{-6}T\right)X_{Na}^2 + \left(0.003 - 5.1\cdot10^{-7}\right)X_{Na}^3 \quad (4)$$

3.2. Surface tension

The new surface tension data obtained for seven ternary liquid $(Sn–Zn)_{eut}$-Na alloys exhibit a linear temperature dependence, and are shown in Fig. 3. The studies pointed at the decrease of surface tension when the concentration and temperature increase, so the surface tension of $(SnZn)_{eut}$-Na reaches the lowest value for the maximal composition of Na (5% (at. %)) The addition of Na to $(SnZn)_{eut}$ should reduce the surface tension, because this (192 mN*m^{-1} at T = 298 K [24]) is lower than the surface tensions for Sn and Zn. Sodium also improves the wettability [25], most probably by removing the oxygen absorbed at the surface of soldered elements (due to the high chemical affinity of Na to O_2). Comparative studies of the surface tension were conducted using