A R C H I V E S O F M E T A L L U R G Y A N D M A T E R I A L S
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TRENDS IN WETTABILITY STUDIES OF Pb-FREE SOLDERS. BASIC AND APPLICATION. PART I. SURFACE TENSION AND DENSITY MEASUREMENTS OF Sn-Zn AND Sn-Zn-Bi-Sb ALLOYS. EXPERIMENT VS. MODELING

KIERUNKI BADAŃ ZWILŻALNOŚCI LUTOWI BEZOŁOWIOWYCH. BADANIA PODSTAWOWE I APLIKACYJNE. CZĘŚĆ I. POMIARY NAPIĘCIA POWIERZCHNIOWEGO I GĘSTOŚCI STOPÓW Sn-Zn I Sn-Zn-Bi-Sb. EKSPERYMENT VS. MODELOWANIE

Surface tension and density measurements by maximum bubble pressure and dilatometric techniques were carried out in an extensive range of temperature on liquid alloys close to binary eutectic Sn-Zn composition with small Bi and Sb additions. It has been found that the addition of Bi and Sb decreases the surface tension and density. The values of the surface tension were also calculated by the Butler model using ADAMIS database from Tohoku University and this of COST 531 program for excess Gibbs energies of liquid components. For modeling, surface tension and density of pure components from SURDAT database were accepted . Calculated surface tension is close to experimental results. Values at 523 K were compared with results from meniscographic/wetting balance studies undertaken in cooperation with industrial institutes in the frame of net-work: "Advanced soldering materials" financed by Polish sources 2006/2007.

The main aim of these joint studies combining basic research with application is to search for substitute materials of traditional solders based on Sn-Pb eutectics and to propose the metric of wettability suitable for different Pb-free materials.

Pomiary napięcia powierzchniowego i gęstości zostały wykonane w znacznym zakresie temperatur dla ciekłych stopów o składzie bliskim eutektyki Sn-Zn z małymi dodatkami Bi i Sb metodą maksymalnego ciśnienia w pcherzykach gazowych i techniką dylatometryczną. Wykazano, że dodatek Bi i Sb obniża napięcie powierzchniowe i gęstość. Przeprowadzono również obliczenia napięcia powierzchniowego za pomoca modelu Butlera używając danych resztkowych energii Gibbsa ciekłych składników z bazy danych ADAMIS z Uniwersytetu w Tohoku oraz z programu COST 531. Dla modelowania, wielkości napięcia powierzchniowego i gęstości czystych składników zostały zaczerpnięte z bazy danych SURDAT. Obliczone wielkości napięcia powierzchniowego są bliskie danym doświadczalnym. Wartości napięcia powierzchniowego w 523 K zostały porównane z wynikami meniskograficznymi w ramach współpracy z instytutami przemysłowymi w sieci: „Zaawansowane materiały lutownicze" finansowanej przez Polską stronę.

1. Introduction

Solderability that is dependent upon wettability of two surfaces being joined is crucial to the efficiency of manufacturing and the reliability of electronic devices. Wettability is generally described by the contact angle, ϕ , to the substrate existing in Young-Dupre relationship. Contact angle as described previously [1] may be calculated from meniscographic studies or directly obtained by the sessile drop method.

Improvement of wettability by lowering of the surface tension was the basic information in the preliminary studies when modeling new solders. This was moderately easy by the Butler's method basing on thermodynamic data of the components of the investigated materials and the surface tension of pure components. However, in the proceeding years it became clear that the spectrum of substitute materials is very limited and studies of different properties, including wettability, are required. This was the starting point to initiate in 1998, at the Institute of Metallurgy and Materials Science (IMIM PAS) in Krakow, the systematic measurements of the surface tension and density of tin with various additions replacing lead . Results of these measurements, carried out by means of the maximum bubble method and dilatometric technique at higher temperatures, were extrapolated to

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250°C to make the comparison with results of meniscographic studies from ITR from Warszawa of interfacial tension in addition to wetting time and wetting force. The latter enabled calculations of contact angles from interfacial tension and wetting force. Thus in the joint studies of wettability on Cu substrate two groups of new Pb-free solders were tested. The first one was based on Sn-Ag and Sn-Ag-Cu (SAC) eutectics, while the second, initiated in recent years, attempted to adopt Sn-Zn eutectics. Due to poorer wettability and higher melting temperature in comparison to Sn-Pb eutectics, both groups require studies on addition of various components to reach the properties closer to traditional materials. In the case of studies starting from Sn-Ag liquid alloys [1] data of surface tension and density including also eutectic composition were used for comparison with wetting balance results when examining two alloys close to ternary eutectic Sn-Ag-Cu (Sn2.76Ag0.46Cu, Sn3.13Ag0.74Cu) [1]. Also the higher amount of Cu added to binary eutectics Sn3.8Ag were tested to eliminate these alloys due to increasing tendency of surface tension [2]. Two wetting balance testers were used, one for measurements of wetting force and wetting time using Cu samples and flux, and the second with non-wetting samples to get solder/flux interfacial tension using Miyazaki method [3]. The wetting force obtained from latter one enables calculations of contact angles. Using the same solders, the same tendency of lowering of the surface tension from maximum bubble pressure method and interfacial tension from wetting balance studies was observed. This is documented in Fig.1, clearly indicating higher values in comparison with eutectic Sn-Pb solder.

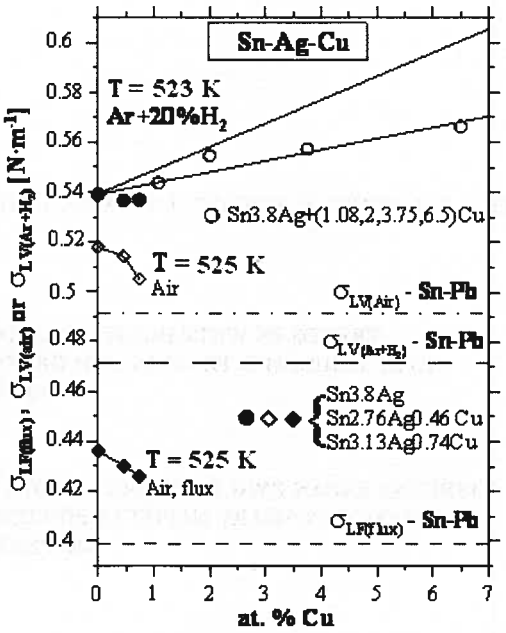

Fig. 1. Effect of Cu additions to Sn-Ag eutectic alloy on surface and interfacial tension

Upper and middle plots show results of maximum bubble pressure measurements of surface tension. Upper plot presents results obtained under protective atmosphere while middle plot – the results obtained in the air from the Miyazaki method.[3]. The lower plot represents the solder/flux interfacial tension from meniscographic method. The open circles in the upper plot are older data [2], which are included to show that the initial addition of Cu first decreases the surface tension from that of the eutectic Sn-Ag solder toward that of the Sn-Ag-Cu ternary eutectic but it rises again on the Cu-rich side of the ternary eutectic. Corresponding invariant values for classic Sn-Pb solders are shown by horizontal dashed or dash-dot lines for comparison. The values of interfacial tension lower than surface tension are due to the flux application.

As the amount of Cu in SAC is limited, and the surface tension should be as close to this of Sn-Pb eutectics as possible, joint studies were initiated on Bi and Sb additions to previously tested ternary alloys. The alloys prepared and analyzed in INMET IMN were studied using the techniques from IMIM PAS and ITR. In addition, studies of electrical and mechanical properties were conducted [4]. A series of three publications [1,5,6] together with another one based on presentation during TMS 134th Annual Meeting @ Exhibition in 2005 [7] (in preparation) present the results of extensive studies on Sn-Ag-Cu-Bi and Sn-Ag-Cu-Bi-Sb alloys with various additions of Bi and Sb. The following conclusions were drawn from the analysis of the data at the temperature 250°C, which is usually used in industrial applications of Pb-free solders:

1. In Sn-Ag-Cu-Bi alloys it was shown that the combination of the results of surface tension with interfacial tensions (measured in air) and the calculated contact angles is the most important as a metric of wettability. The changes of these parameters correlate with decreasing wetting time and increasing wetting force simultaneously with decreasing surface tension (from maximum bubble pressure method).

2. Similar results on quinary Sn-Ag-Cu-Bi-Sb [7] alloys do not show a continuation of these trends.

Understanding and resolving of this problem became possible with the suggestions concerning the role of wetting time and wetting force in a recent paper of Lopez et al. [8]. In Ref. [8] it was indicated that wetting force is not a generalized metric of wettability because it cannot account for the significant effect of solder/flux interfacial tension on the wetting and spreading phenomenon. On the other hand, wetting time is rather more representative of wetting kinetics than wettability. Because of this, the wetting time and wetting force together with contact angles are often used in practice for comparison of various solders, but they do not exhibit a general dependence upon composition as does the interfacial tension in combination with contact angles or the surface tension (from the maximum bubble pressure method) in combination with contact angles. Refs. [5-7] present the combination of the results of wettability studies with calculations of phase equilibria in the alloys with both Bi and Sb. The studies were carried out in cooperation with Tohoku University in Japan. It was shown that only the small amounts of both additions are useful, as the higher amounts result in fillet lifting effect due to the segregation of Bi at the solder/substrate boundary.

In these studies, we intend to test the positive influence of Bi and Sb indicated in previous publications [5-7] in wettability of Sn-Zn-Bi-Sb alloys, starting from Sn-Zn eutectic. It has been also shown previously that the addition of Sb to Sn, to binary eutectic Sn-Ag and to Sn3.3Ag0.74Cu decreases the surface tension and density [9]. The latter conclusions resulted from studies within COST 531 Program in which also additions of In to binary and ternary Sn-Ag and Sn-Ag-Cu alloys were tested [10]. Examining the In additions in systems in which simultaneously Sn is present like In-Sn, Ag-Sn-In, Sn-Cu-Ag-In due to the nearly the same surface tension and density of pure indium and tin in alloys, no evident concentration dependence was observed. Positive influence of In on wettability, documented in previous Refs. [11-12] was indicated by the lowering of the contact angle. The same was true, when discussing the results of In additions to Sn-Zn alloys in doctor's thesis of J. Pstruś [13]. In Ref. [10] in addition to the surface tension and contact angles, the influence of In (0-75 at.%) additions to the Sn3.13Ag0.74Cu solder on the structure of the interface between relevant solder and Cu for various temperatures (250, 280 and 320°C) were investigated. Accepting the importance of the contact angle, it seems interesting to test how other factors connected with wettability like surface tension, interfacial tension, wetting time and wetting force are changed in the case of various Pb-free solders.

Studies of the addition of small amounts of Bi and Sb to eutectic Sn-Zn will be divided into two parts. In Part I, the measurements of surface tension and density in protective atmosphere of Ar + H_2 will be undertaken and compared with modeling by the Butler's model. In Part II, surface tension measurements will be compared with results from wetting balance studies both in protective atmosphere and in air, and verified by wettability on Cu substrate with the use of various fluxes.

Systematic studies of surface tension, density and modeling of surface tension of the substitute materials for traditional Sn-Pb solders were initiated in 1998 in IMIM PAS. They were used in preparations of SURDAT database published in 2007 [14] and distributed to various centers including COST 531 Program (2002-2006) and international net-work ELFNET (2004-2006).

2. Surface tension measurements of Sn-Zn 16.7 and Sn-Zn 12.2 with Bi and Sb additions

Wettability studies undertaken jointly in IMIM PAS and ITR were conducted on the same alloys prepared and analyzed in INMET (Institute of Non-Ferrous Metallurgy in Gliwice).

Two kinds of starting alloys, Sn-Zn with the same amount of Bi and Sb were prepared according to the information from literature [15], which suggested the use of materials with lower amount of Zn than in eutectic alloy. This lowers the tendency to oxidation and probably it is easier to find the suitable flux. In this Part 1, composition of alloys are given in atomic percent. However in Part 2 the mass % were used, as in the industrial practice, and therefore in Tables 1 and 2 both atomic and mass % are given.

TABLE 1

Compositions of alloys with 16.7 atomic % Zn

Composition Mass %				Composition Atomic %			
Sn	Zn	Bi	Sb	Sn	Zn	Bi	Sb
89.80	9.92	–	–	83.3	16.70		
88.60	9.43	1.17	1.13	82.43	15.92	0.62	1.03
87.30	9.55	2.20	1.18	81.56	16.20	1.17	1.07
86.80	9.54	1.08	2.45	81.03	16.17	0.57	2.23
87.00	9.03	2.27	2.17	81.46	15.35	1.21	1.98

TABLE 2

Compositions of alloys with 12.2 atomic % Zn

Composition Mass %				Composition Atomic %			
Sn	Zn	Bi	Sb	Sn	Zn	Bi	Sb
92.92	7.08			87.8	12.2		
90.70	6.99	1.07	1.24	86.21	12.06	0.58	1.15
89.50	7.00	2.25	1.25	85.48	12.14	1.22	1.16
90.00	6.76	1.06	2.18	85.71	11.69	0.57	2.02
88.60	6.90	2.30	2.20	84.72	11.98	1.25	2.05

In a surface tension measurements at the extensive range of temperature the maximum bubble pressure method in protective atmosphere argon + hydrogen was used. Experimental details were presented in earlier publication [1]. Results of the surface tension measurements are summarized in Tables 3 and 4, and plotted in Figures 2 and 3.

TABLE 3

Temperature dependencies of the surface tension of the liquid binary Sn-Zn16.7 and quaternary Sn-Zn-Bi-Sb alloys with the calculated errors of the A and B parameters and the surface tension values calculated at 523 K

Composition Atomic %				σ = A + B*T(K) mN/m	σ (523 K) mN/m	Err(A) mN/m	Err(B) mN/m K
Sn	Zn	Bi	Sb				
83.3	16.70			586.9 – 0.0673	552 ± 8	± 11.4	± 0.0141
82.43	15.92	0.62	1.03	579.6 – 0.0659	545 ± 4	± 7.5	± 0.0105
81.56	16.20	1.17	1.07	579.6 – 0.0687	544 ± 8	± 11.2	± 0.0138
81.03	16.17	0.57	2.23	577.6 – 0.0621	545 ± 4	± 5.2	± 0.0064
81.46	15.35	1.21	1.98	568.3 – 0.0650	534 ± 4	± 6.4	± 0.0087

TABLE 4

Temperature dependencies of the surface tension of liquid binary Sn-Zn12.2 and quaternary Sn-Zn-Bi-Sb alloys with the calculated errors of the A and B parameters and the surface tension values calculated at 523 K

Composition Atomic %				σ = A + B*T(K) mN/m	σ (523 K) mN/m	Err(A) mN/m	Err(B) mN/m K
Sn	Zn	Bi	Sb				
87.8	12.2			584.8 – 0.0675	550 ± 3.6	± 5.2	± 0.0069
86.21	12.06	0.58	1.15	571.1 – 0.0556	542 ± 6	± 10.0	± 0.0145
85.48	12.14	1.22	1.16	565.2 – 0.05137	538 ± 3	± 3.4	± 0.0047
85.71	11.69	0.57	2.02	569.0 – 0.0608	569 ± 3	± 4.1	± 0.0057
84.72	11.98	1.25	2.05	569.8 – 0.0713	533 ± 3	± 4.6	± 0.0062

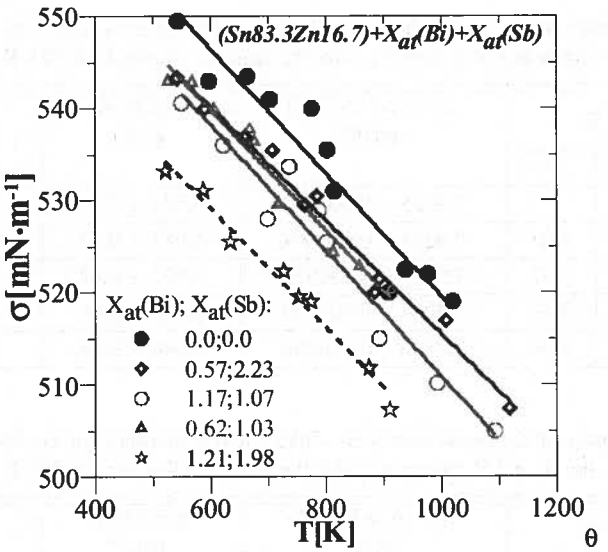

Fig. 2. Temperature dependencies of the surface tension of the liquid binary Sn-Zn16.7 and quaternary Sn-Zn-Bi-Sb

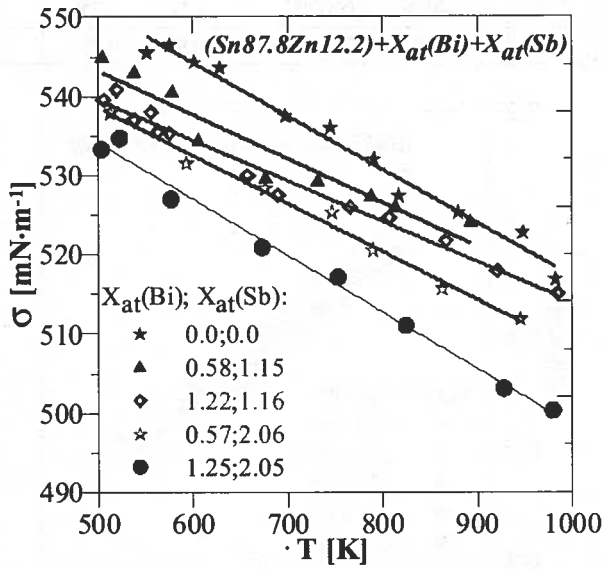

Fig. 3. Temperature dependencies of the surface tension of the liquid quaternary Sn-Zn-Bi-Sb alloys based on Sn-Zn12.2

It is evident from Fig.2 and Fig.3 that the results of surface tension of both alloys Sn-Zn are lower than for quaternary alloys Sn-Zn-Bi-Sb, and the lowest values of surface tension are for highest content of Bi and Sb. Values of surface tension of quaternary alloys at 523 K will be compared and analyzed with results from wetting balance data and with calculated results from Butler's model in Part II.

3. Density measurements of Sn-Zn16.7 and Sn-Zn 12.2 with Bi and Sb additions

Density measurements were performed by the dilatometric method for the same alloys and temperatures as previously described in surface tension studies. Data of densities are presented in Tables 5 and 6, while Figs. 4 and 5 represent results at the entire range of temperature for both group of alloys based on Sn- Zn.

TABLE 5

Temperature dependencies of the density of the liquid binary Sn-Zn16.7 and quaternary Sn-Zn-Bi-Sb alloys with the calculated errors of the A and B parameters and the density calculated at 523 K

Composition Atomic %				σ = A + B*T(K) g/cm^3	ρ (523 K) g/cm^3	Err(A) g/cm^3	Err(B) g/cm^{3}K
Sn	Zn	Bi	Sb				
83.3	16.70			7.3525 – 0.000729	6.971 ± 0.06	± 0.079	± 0.000094
82.43	15.92	0.62	1.03	7.4334 – 0.000864	6.982 ± 0.03	± 0.065	± 0.000083
81.56	16.20	1.17	1.07	7.3331 – 0.000678	6.979 ± 0.02	± 0.034	± 0.000045
81.03	16.17	0.57	2.23	7.4034 – 0.000790	6.990 ± 0.06	± 0.096	± 0.000121
81.46	15.35	1.21	1.98	7.4640 – 0.000799	7.046 ± 0.08	± 0.105	± 0.000137

TABLE 6

Temperature dependencies of the density of the liquid binary Sn-Zn12.2 and quaternary Sn-Zn-Bi-Sb alloys with the calculated errors of the A and B parameters and the density calculated at 523 K

Composition Atomic %				σ = A + B*T(K) g/cm^3	ρ (523 K) g/cm^3	Err(A) g/cm^3	Err(B) g/cm^{3}K
Sn	Zn	Bi	Sb				
87.8	12.2			7.3565 – 0.000709	6. 986 ± 0.05	± 0.062	± 0.000089
86,21	12,06	0,58	1,15	7,3463 – 0,000715	6,972 ± 0,02	± 0,045	± 0,000065
85,48	12,14	1,22	1,16	7,3999 – 0,000733	7,016 ± 0,05	± 0,104	± 0,000162
85,71	11,69	0,57	2,02	7,4488 – 0,000797	7,032 ± 0,04	± 0,066	± 0,000100
84,72	11,98	1,25	2,05	7,3425 – 0,000690	6,982 ± 0,03	± 0,065	± 0,000099

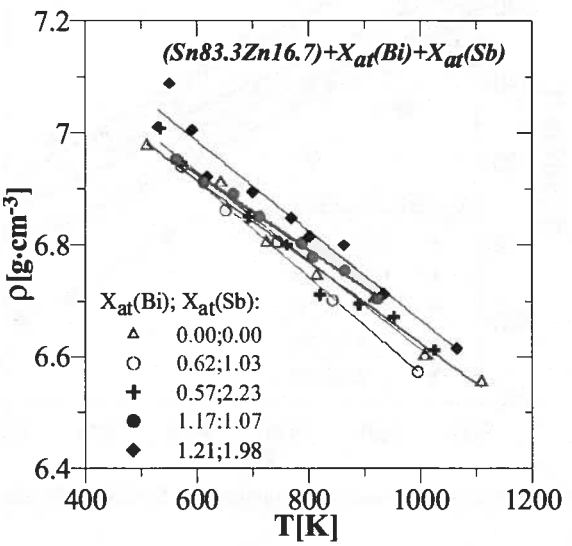

Fig. 4. Temperature dependencies of the density of the liquid binary Sn-Zn16.7 and quaternary Sn-Zn-Bi-Sb alloys

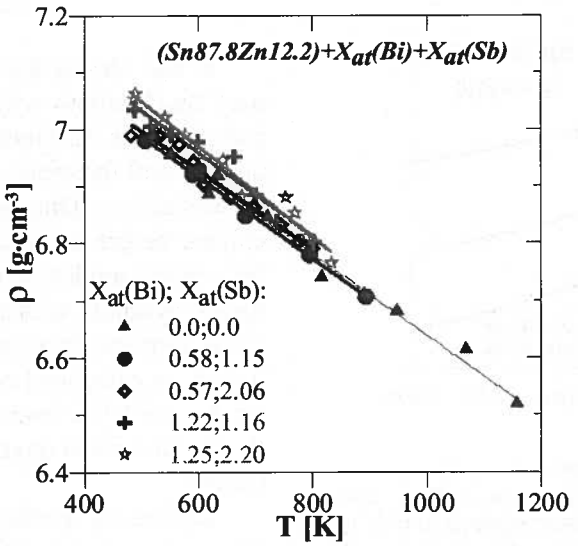

Fig. 5. Temperature dependencies of the density of the liquid binary Sn-Zn12.2 and quaternary Sn-Zn-Bi-Sb alloys

Data of densities are needed for calculations in wetting balance studies and for modeling of the surface tension. New data of surface tension and density will be introduced also into the updated SURDAT database.

4. Modeling of surface tension of Sn-Zn-Bi-Sb alloys

To model the surface tension Butler's method was used [17]. This method requires data of the excess Gibbs energies of the components of the bulk phase of the investigated liquid alloys, and the values of the surface tension of pure components. In modeling of surface tension of liquid Sn-Zn-Bi-Sb alloys three databases were used. Density and surface tension of pure components Sn, Zn, Bi and Sb were accepted from SURDAT database [14].

Data of excess Gibbs energies of liquid components of Sn-Zn-Bi-Sb alloys were calculated basing on information from constituent binary data, and in addition the interaction parameters of two known ternary systems Bi-Sb-Sn and Bi-Sn-Zn were taken into consideration. These data were stored in ADAMIS [18] and COST 531 thermodynamic databases.

Calculations of the surface tension were performed at several temperatures including 523 K usually accepted for comparison with wetting balance results, which will be discussed in Part II of these joint studies. Results of calculations in molar fractions are shown in Figures 6-7 to indicate the influence of Bi additions, and in Figures 8-9 to show the effect of Sb additions. The Sn-Zn (0.833Sn0.167Zn) eutectic alloy was always the starting point.

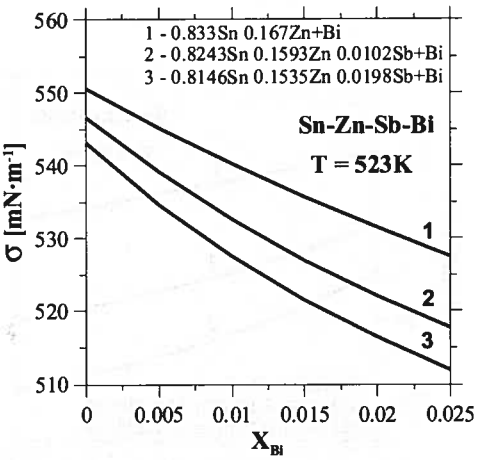

Fig. 6. Calculated by Butler's model influence of Bi additions on surface tension of binary alloy 0.833Sn0.167Zn and on ternary alloys containing 0.8243Sn0.1593Zn0.0102Sb, 0.8146Sn0.1535Zn0.0198Sb molar fractions of Sn, Zn and Sb at 523 K

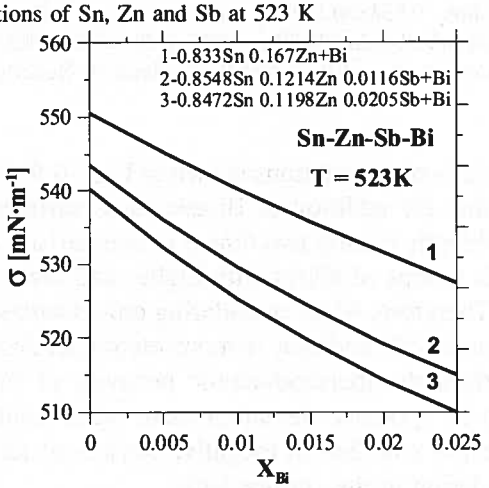

Fig. 7. The influence of Bi additions on surface tension of binary alloy 0.833Sn0.167Zn and on ternary alloys containing 0.8548Sn0.1214Zn0.0116Sb, 0.8472Sn0.1198Zn0.0205Sb molar fractions of Sn, Zn and Sb at 523 K calculated by Butler's model

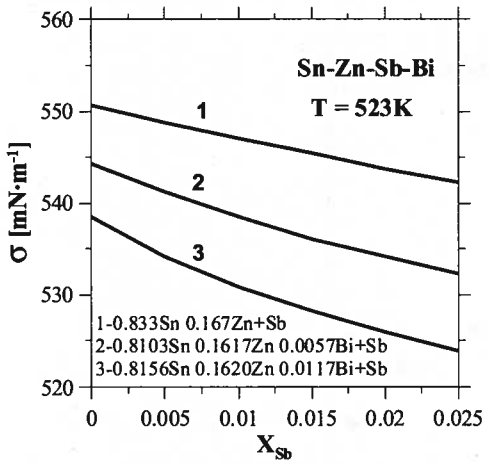

Fig. 8. The influence of Sb additions on surface tension of binary alloy 0.833Sn0.167Zn and on ternary alloys containing 0.8103Sn0.1617Zn0.0057Bi, 0.8156Sn0.1620Zn0.0117Bi molar fractions of Sn, Zn and Bi at 523 K calculated by Butler's model

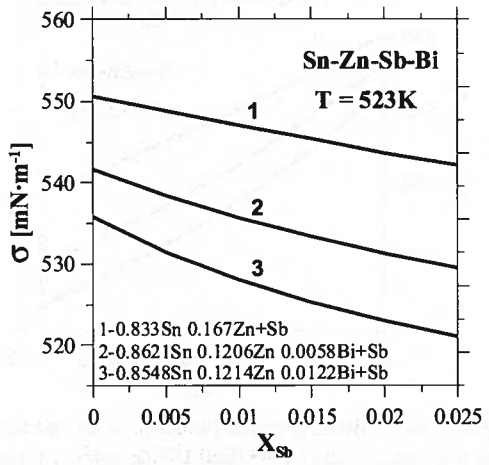

Fig. 9. The influence of Sb additions on surface tension of binary alloy 0.833Sn0.167Zn and on ternary alloys containing 0.8621Sn0.1206Zn0.0058Bi, 0.8571Sn0.1169Zn0.0057Bi molar fractions of Sn, Zn and Bi at 523 K calculated by Butler's model

Results of calculations shown in Figs. 6-9 clearly indicate that the addition of Bi decreases surface tension very evidently (nearly two times) in comparison with Sb, for both groups of alloys with higher and lower amount of Zn. Therefore, when considering only decrease of surface tension, Bi addition is more recommended. It may result from the thermodynamic behavior of binary alloys Sn-Bi (positive deviation from ideal solutions) in comparison with Sn-Sb (negative deviation) and the Bi accumulation in the surface layer.

The calculated values of surface tension of quaternary alloys Sn-Zn-Bi-Sb are very close to the experimental data presented in Tables 3 and 4 and plotted in Figs. 2 and 3.

5. Conclusions

It was shown that the addition of Bi and Sb to binary Sn-Zn alloys with 9.9 mass % Zn (nearly eutectic) and of lower Zn amount decreases surface tension and density and therefore should have the positive influence on wettability. One has to be anxious with such conclusion before meniscografic studies and calculation of the contact angles. It will be shown in Part II based on meniscographic measurements with the use of various fluxes that the decrease of interfacial tension is not sufficient, as calculated contact angles for starting eutectics Sn-Zn are lower than for Sn-Zn-Bi-Sb. This statement finally eliminates quaternary alloys from practical applications.

Basing on positive influence of Bi and Sb additions on surface tension from maximum bubble pressure method and on the same indication from wetting balance studies together with the calculated values by Butler's method, the thermodynamic studies on quaternary alloys were initiated. This was done by means of high temperature galvanic cells for constituent ternaries and for quaternary Zn-Sn-Bi-Sb alloys together with DTA. Thus the data for phase diagram calculations were prepared. This will be presented in Part III.

When the cooperation program of IMIM PAS with industrial institutes ITR and INMET IMN, and simultaneously with Tohoku University has been started at 2000, the search of the metric of wettability seemed much simpler than nowadays. Each new alloy intended to substitute the traditional Sn-Pb solder requires special studies starting from SURDAT database of surface tension, interfacial tension (this will be added in updated version) and contact angle calculated or measured. In these attempts it seems useful to combine the basic studies with the practice of industry and to continue the joint efforts, taking into account that the process of introducing Pb-free alloys initiated more than 10 years ago is not completed and a new substitute material was not proposed with the equivalent properties to Sn-Pb solder.

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