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Chapter

Surface Tension and Surface Tension Assessment of Ag-Au-Cu Ternary and Sub-Binary Alloy Systems

Hüseyin Arslan and Ali Dogan

Abstract

A brief review of measurement techniques and theoretical studies on the surface tension alloy and mixture has been presented in the present study. It is clear that the experimental determination of thermodynamic and thermophysical properties of both solid and especially liquid alloys at high temperature cases is frequently difficult technologically. In addition to this, a lack of experimental data concerning thermophysical properties of Ag-Au, Au-Cu, and Ag-Cu sub-binary systems is obvious. The theoretical thermophysical data of the Ag-Au-Cu ternary alloy systems are very scarce in the literature. Thus, the surface tensions of the alloys just mentioned above for cross sections $z = x_{Ag}/x_{Au} = 1/3, 1/1, 3/1, 2/5$, and $5/2$, respectively, and their sub-binary systems are much simply calculated from the surface tensions of the Ag-Au, Au-Cu, and Ag-Cu sub-binary systems by using geometric models, such as Muggianu, Kohler, Toop, and GSM (Chou's general solution model) and Butler's equation. The predicted results in the present study show rather an agreement with the experimental results of the alloys. Therefore, it is inferred that the obtained surface tension curves for the Ag-Au-Cu ternary alloy at 1381 K are reasonable with especially those calculated from the Toop model.

Keywords: surface tension, geometric models, Butler equation, Pb-free ternary alloys

1. Introduction

It is seen that the surface tensions of the materials are of outstanding importance from many scientific and technological viewpoints. Surface tensions have been measured for a long time and it is seen that the collections of experimental data for pure liquids [1–11] and some binary liquid alloy systems exist. Surface tension measuring techniques can be classified generally as goniometric and tensiometric. They can also be classified into two classes. The first class is static surface tension measurements. When the values of surface tension are constant, the pure liquids are measured with these devices. The second one is dynamic surface tension measurements so that many of these are considered as the modifications of the static models. One can generally mention some experimental methods such as ring [1, 12]; oscillating jet [13–17]; DC method, as described in detail elsewhere [18–20]; oscillating

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droplet method [21–40]; draining crucible method [19, 20, 41, 42]; drop or weight method (it can be seen that the drop volume or weight method among the conventional methods of surface tension measurement has proven to be reliable and easy to handle) [43–47]; pulsating bubble [48]; pendant drop (it may be said that the use of the pendant drop method to measure interfacial tension between molten polymers has gotten a lot of attention) [11, 49–53]; sessile drop [54–78]; and maximum bubble pressure methods (this method is one of the most popular techniques to measure the dynamic surface tension of various surfactants).

This method is particularly useful in measuring surface tension of highly concentrated surfactant solutions [79] and molten metals [80–83] for binary alloys [84–90] for multicomponent alloys like Au-based [87, 91–106]. The last one among these methods has received much attention recently.

Recently, some researchers [19, 20, 41] developed a new method for fluids to simultaneously measure the surface tension, viscosity, and density in only one experiment.

Although a brief review of the experiments is given above, it is impossible to say that the experiments carried out are sufficient. On the other hand, the surface tension prediction is useful in designing and discovering new materials and it is necessary to discuss them theoretically. A brief review of some theoretical studies can be given here [16, 33, 71, 101, 104, 107–159] along with neural network modeling dealing with the alloys and mixtures. The artificial neural network (ANN) studies have been carried out to predict the surface tension of some chemicals including liquid drugs [160] and the alloys Sn-In-Zn-Ag using Butler model, rare earth containing binary chloride mixtures via STCBE computer program [161], and the binary alloys Fe-Cu, Cu-Pb, Sn-Pb, Ag-Pb, Pb-In, Bi-Ag, Ag-Sn, Cu-A1, Fe-Si, and Ni-Si via a special calculation technique [135, 162]. In addition, the surface tensions of the binary alloys in some low melting metal systems (Pb-Sb, Ag-Bi, Ag-In, Ag-Sn, Bi-Sn, and Sb-Sn) using a thermodynamic database and a database of densities and surface tensions of the respective pure metals based on published experimental data are given in ref. [163].

It is impossible to avoid traces of oxygen in the working atmosphere. In such situations, the surface contamination phenomena continues to be important in determining reliable thermophysical properties, such as data of surface tension, density, etc. Recently, for high-temperature measurements and studies of reactive materials, the containerless processing techniques in the experimental studies carried out have been used in order to isolate samples from their environment and to reduce contamination [164]. The surface tensions of liquid metals, Zr, Ni, Ti, Mo, and Nb, have been measured at their melting points using the quasi-containerless pendant drop method, which is much reliable for contamination [3]. Some critical investigations revealed that only a small number of papers concerning systematic investigations of surface tension of the multicomponent systems have been published on the effect of a wide range of temperature and concentration on the thermophysical properties. It can also be seen that the effect of a wide range of temperature and concentration on the surface tension of liquid ternary alloy systems is very sparse. Recently, the surface tension data for the Ag-Au, Au-Cu, and Ag-Cu binary alloys have been measured and reported in literature by some authors [165–169]. However, to the best of our knowledge, there is disagreement between data from different authors concerning the existence with agreements in the selected surface tensions of the components Ag, Au, and Cu. When an analytical expression of γ_i, experimental data, the surface tensions of the Ag-Au [90], Au-Cu [170], and Ag-Cu [59] binary alloys at 1381 K by using Muggianu model [171] were calculated and those of the Ag-Au-Cu ternary alloy at 1381 K by using geometric models, such as Kohler [172], Toop [173], GSM [174], and Butler's equation [175],

were compared with the experimental data measured by Pajarre et al. [167] and Gallois and Lupis [166] in the present study. Hu et al. [168] first extended geometric models to predict the surface tensions of the Ag-Au-Cu ternary alloy but did not make a comparison between the calculated and experimental results. In the present study, the case in question was carried out and the ratios of silver/gold were kept constant, and the amount of copper was systematically changed with the ratio $z = x_{Ag}/x_{Au}$ = 1/3, 1/1, 3/1, 2/5, and 5/2, respectively while computing the surface tensions. Moreover, the symmetry properties of the Ag-Au-Cu ternary alloy system were judged. On the other hand, the excess surface tensions instead of excess thermodynamic properties encountered frequently in researches calculating the Gibbs energies and enthalpies of the alloys [176–183] have been used in order to calculate the surface tension of related ternary alloys in the present study [2, 46, 62, 86, 97, 100, 174, 176–181, 184–188].

2. Background of excess and ideal surface tension and geometric models

Excess surface tension (σ^E) has been calculated by Eq. (1):

$$\sigma^E = \sigma - \sigma^i \tag{1}$$

where

$$\sigma^i = \sum_{i=1}^{2} x_i \sigma_i \tag{2}$$

here, x_i and σ_i are the mole fraction and surface tension of the pure liquid component i.

A thermodynamic property of mixing, such as surface tension, can be expressed as a combination of all binaries with an assigned probability weight and the thermodynamic properties of a ternary system can be then calculated by

$$\sigma^E = \sum_{ij} W_{ij} \sigma_{ij}^E, \ (i \neq j = 1 \text{ to } 3) \tag{3}$$

where W_{ij} represents the weight probability of each corresponding binary composition point and can be calculated via:

$$W_{ij} = \frac{x_i x_j}{X_{i(ij)} X_{j(ij)}} \tag{4}$$

where $X_{i(ij)}$ and $X_{j(ij)}$ indicate the mole fractions of component i and j in the binary system and are given different forms according to the selected geometric models.

The excess surface tension in Eq. (3) associated with the binary subsystems of the alloy systems is given in the form of Redlich-Kister polynomials:

$$\sigma^E{}_{ij} = x_i x_j \left[\left(A_{ij}^0 + B_{ij}^0 T \right) + \left(A_{ij}^1 + B_{ij}^1 T \right) \left(x_i - x_j \right) + \left(A_{ij}^2 + B_{ij}^2 T \right) \left(x_i - x_j \right)^2 + \cdots \right] \tag{5}$$

The Muggianu model can be expressed readily in a series form so that A_{ij} denotes the Redlich-Kister parameters:

$$\sigma^E = \sum_{i=1}^{2}\sum_{j>i}^{3} x_i x_j A_{ij} + x_i\, x_{j>i}\, x_{k>j}\, A_{i,j,k} \tag{6}$$

where

$$A_{ij} = \sum_{\vartheta=0}^{2} A_{ij}^{\vartheta}\left(x_i - x_j\right)^{\vartheta} \tag{7}$$

$$A_{i,j,k} = x_i A_{i,j,k}^{0} + x_j A_{i,j,k}^{1} + x_k A_{i,j,k}^{2} \tag{8}$$

The Kohler model can be given in a closed form simply:

$$\begin{aligned}\sigma^E = {} & (x_1+x_2)^2\sigma^E{}_{12}\left(\frac{x_1}{x_1+x_2};\frac{x_2}{x_1+x_2}\right) + (x_3+x_1)^2\sigma^E{}_{31}\left(\frac{x_3}{x_1+x_3};\frac{x_1}{x_1+x_3}\right) \\ & + (x_2+x_3)^2\sigma^E{}_{23}\left(\frac{x_2}{x_2+x_3};\frac{x_3}{x_2+x_3}\right)\end{aligned} \tag{9}$$

The Toop model can also be given in a closed form simply:

$$\sigma^E = \frac{x_2}{1-x_1}\sigma^E{}_{12}(x_1;1-x_1) + \frac{x_3}{1-x_1}\sigma^E{}_{13}(x_1;1-x_1) + (x_2+x_3)^2\sigma^E{}_{23}\left(\frac{x_2}{x_2+x_3};\frac{x_3}{x_2+x_3}\right) \tag{10}$$

Using Eqs. (3) and (4), the expression of the surface tension concerning the GSM model can be written as:

$$\sigma^E = \frac{X_1X_2}{X_{1(12)}X_{2(12)}}\sigma^E{}_{12} + \frac{X_2X_3}{X_{2(23)}X_{3(23)}}\sigma^E{}_{23} + \frac{X_3X_1}{X_{3(31)}X_{1(31)}}\sigma^E{}_{31} \tag{11}$$

This model gets rid of the asymmetric component selecting problems seen in the Toop model. The compositions of binary alloys in Eq. (11) can be written as:

$$X_{1(12)} = X_1 + X_3\xi_{12} \tag{12}$$

$$X_{2(23)} = X_2 + X_1\xi_{23} \tag{13}$$

$$X_{3(31)} = X_3 + X_2\xi_{31} \tag{14}$$

The key step in the GSM is introducing a new concept, called a similarity coefficient, ξ, into the geometrical model, that is,

$$\xi_{ij}^{k} = \frac{\eta(ij,ik)}{\eta(ij,ik)+\eta(ji,jk)} \tag{15}$$

where $\eta(ij, ik)$ is a function related to the excess Gibbs free energy of ij and ik two binaries and is given as follows:

$$\begin{aligned}\eta(ij,ik) = \int_{x_i-0}^{x_i-1}\left(\sigma_{ij}^E - \sigma_{ik}^E\right)^2 dX_i = \eta(ij,ik) = {} & \sum_{l=0}^{n}\frac{1}{2(2l+1)(2l+3)(2l+5)}\left(A_{ij}^l - A_{ik}^l\right)^2 \\ & + \sum_{l=0}^{n}\sum_{m>l}^{n}\frac{1}{(l+m+1)(l+m+3)(l+m+5)} \times \left(A_{ij}^l - A_{ik}^l\right)\left(A_{ij}^m - A_{ik}^m\right)\end{aligned} \tag{16}$$

Its value up to three order of k, for instance, can be given as follows readily:

$$\eta(12,13) = \frac{1}{30}\left(A_{12}^{0} - A_{13}^{0}\right)^{2} + \frac{1}{210}\left(A_{12}^{1} - A_{13}^{1}\right)^{2} + \frac{1}{630}\left(A_{12}^{2} - A_{13}^{2}\right)^{2} + \frac{1}{105}\left(A_{12}^{0} - A_{13}^{0}\right)\left(A_{12}^{2} - A_{13}^{2}\right) \tag{17}$$

In addition, it should be noted that $A_{ij}{}^{k} = A_{ji}{}^{k}$, when k is even and $A_{ij}{}^{k} = -A_{ji}{}^{k}$ and when k is odd in Eqs. (6), (7), (16), and (17).

3. Background of the Butler model

Butler's equation has been used extensively to calculate the surface tension of multicomponent alloy systems. The surface of the bulk is treated as an additional thermodynamic phase, in equilibrium with the bulk phase. If the ternary alloys are taken as a regular solution model, the surface tension of ternary liquid alloys dealing with Butler's model is written as:

$$\begin{aligned} \sigma &= \left\{\sigma_1 + \frac{RT}{S_1}\ln\left(\frac{X_1^s Y_1^s}{X_1^b Y_1^b}\right)\right\} \\ \sigma &= \left\{\sigma_2 + \frac{RT}{S_2}\ln\left(\frac{X_2^s Y_2^s}{X_2^b Y_2^b}\right)\right\} \\ \sigma &= \left\{\sigma_3 + \frac{RT}{S_3}\ln\left(\frac{X_3^s Y_3^s}{X_3^b Y_3^b}\right)\right\} \end{aligned} \tag{18}$$

here, R, T, σ_i, and S_i are gas constant, temperature in terms of K, surface tension of pure component i, and surface area of component i, respectively. The surface area of component i is

$$S_i = 1.091 N_a^{1/3}\left(\frac{M_i}{\rho_i}\right)^{2/3} \tag{19}$$

here, the molar volume of each component V_i can be expressed as M_i/ρ_i. This expression can be calculated from Avogadro's number N_a, the atomic mass M_i, and the density data ρ_i, as in Eq. (19). In this equation, X_j^k represents an alloy composition with the subscript j and superscript k referring to the corresponding component j in the bulk, b, and the surface phase, s. The terms γ_i^b (T, X_j^b) and γ_i^s (T, X_j^s) in Eq. (18) are activity of component i in the bulk phase and the surface phase, respectively. These terms can be obtained as both functions of temperature and composition. The activity term of component i can be derived from standard thermodynamic relationships, in the form:

$$G_i^E = RT \ln \gamma_i = G^E + \sum_{j=1}^{3}\left(\delta_{ij} - x_j\right)\frac{\partial G^E}{\partial X_j} \tag{20}$$

where δ_{ij} is Kronecker's symbol and j = i, $\delta_{i\,j} = 1$ and j ≠ i, $\delta_{i\,j} = 0$. Tanaka et al. [184] have proposed a model for $G_i^{E,s}$ (T, $X_{j\,(j=2;3,\ldots)}^{s}$) as follows:

$$G_i^{E,s}\left(T, X_j^s\right) = \beta\, G_i^{E,b}\left(T, X_j^b\right) \tag{21}$$

where β is a parameter corresponding to the ratio of the coordination number z in the surface phase to that in the bulk phase, z^s/z^b. In some cases, the value of β parameter might be affected by other factors, such as the relaxation of the surface structure, and in the literature, different values between 0.5 and 0.84 are used [62]. The number of nearest neighbors surrounding a central atom is 9 in the surface phase. In addition, the coordination number for the bulk atom is 12. Here, in order to calculate surface tensions of the alloy systems, the value of the parameter β has been taken as 0.83 in the present study.

In order to measure surface tension of the liquids at temperatures above the melting point of alloys on the one hand, the surface tensions of some alloys have been measured contactless using the technique of electromagnetic levitation in combination with the oscillating drop technique [127]. On the other hand, some calculation models have also been developed. In order to calculate surface tension of the binary alloys [170], a new thermodynamic model is proposed by Prasad and Mikula [143]. It is assumed that there is a relation between γ_i,γ_i^s and p ln γ_i^*, q lnγ_i; here, p and q are called as surface coordination fractions in which p = 0.5, q = 0.25 for closed packed structured. When an analytical expression of γ_i is known, it is possible to obtain expressions for γ_i^s, so the surface tension of the binary mixture can be calculated easily. Recently, the surface properties of Ag-Cu and Ag-Ti liquid alloys are predicted by a quasi-chemical solution model (QCS) [189–192]. Whileas the compound formation model (CFM) [162, 193] (it is related with both weak and strong interaction approximation) in Cu-Ti system has been performed. Moreover, the surface tensions of Ag-Ti and Ag-Hf liquid alloys have been predicted by the QCA in the case of regular solutions, developed by Bhatia and Singh [194, 195], in the framework of statistical mechanical theory in conjunction with the quasi-lattice theory, while in the case of the Ag-Ti system, at T = 1773, the CFM has been applied [190, 196].

On the other hand, the value of coefficient f = 1.091 in the surface area of component i in Eq. (19) has been altered by Kaptay and Papp [197]. In addition to this, the model of Butler has been reconstructed from the very beginning to finishing of the model for the surface tension of one-component liquid metals. In their paper, the effects of surface active complexes such as the intermetallic compounds in liquid metallic solutions are also considered.

4. Experimental surface tension data of liquid Ag, Au, and Cu

The surface tensions of liquid Ag, Au, and Cu were measured at 1381 K by Gallois and Lupis [166] under atmospheres of argon, dilute solutions of hydrogen in argon, and under vacuum and are found as σ_{Ag} = 0.890 ± 0.01 (N/m), σ_{Au} = 1.150 ± 0.01 (N/m), and σ_{Cu} = 1.320 ± 0.015 (N/m). Pajarre et al. [167] have used the values σ_{Ag} = 0.892132 [46], σ_{Au} = 1.13666 (N/m), respectively [185], and σ_{Cu} = 1.29499 (N/m) [21] in their experimental and theoretical study in which the experimental surface tension values for the Ag-Au-Cu system have been determined by the sessile drop method at 1381 K [167]. For the surface tension calculations in Surdat, the surface tension data σ_{Ag} = 0.870912 (N/m) [186], σ_{Au} = 1.503 (N/m) [2], and σ_{Cu} = 1.4756 (N/m) [100] are used. As mentioned previously, the surface tension data are very dispersed in the literature. For the calculations carried out in the present study, the values of the surface tension of [166] have been adopted since it is inferred that the experimental results deviate from the corresponding values predicted by the geometric models at the temperature of 1381 K.

System	A_{ij}^{o}	A_{ij}^{1}	A_{ij}^{2}
Ag-Au	−128.79 + 0.045836 T	243.86 − 0.060170 T	−455.82 + 0.185348 T
	+0.00680219 T lnT	−0.00965926 T lnT	+0.02786783 T lnT
	−0.00001833 T^2	+0.00002163 T^2	−0.00011934 T^2
Ag-Cu	−982.68 + 0.262677 T	1625.72 − 0.693760 T	−1142.64 − 0.084365 T
	+0.04028137 T lnT	−0.09772514 T lnT	+0.14883052 T lnT
	−0.00013536 T^2	+ 0.00039080 T^2	−0.00030951 T^2
Au-Cu	94.97 + 0.002865 T	−43.72−0.009243 T	−9.72 − 0.008609 T
	−0.00050101 T lnT	−0.00094336 T lnT	−0.00071243 T lnT
	−0.0000217 T^2	+ 0.00000558 T^2	+ 0.00002569 T^2

Table 1.
Redlich-Kister parameters for excess surface tensions of the Ag-Au, Ag-Cu, and Au-Cu sub-binary alloys at 1381 K.

By using the values of Redlich-Kister parameters of the binary alloys given in references [2, 86, 97] and the surface tensions of liquid Ag, Au, and Cu measured at 1381 K in the reference [166], the excess binary surface tensions σE and binary surface tensions σ of three sub-binary systems are calculated and shown in **Figure 1**. Using Eqs. (1)–(4) and the other equations relevant to the models, the surface tension reference data for the pure components, such as the surface tension of

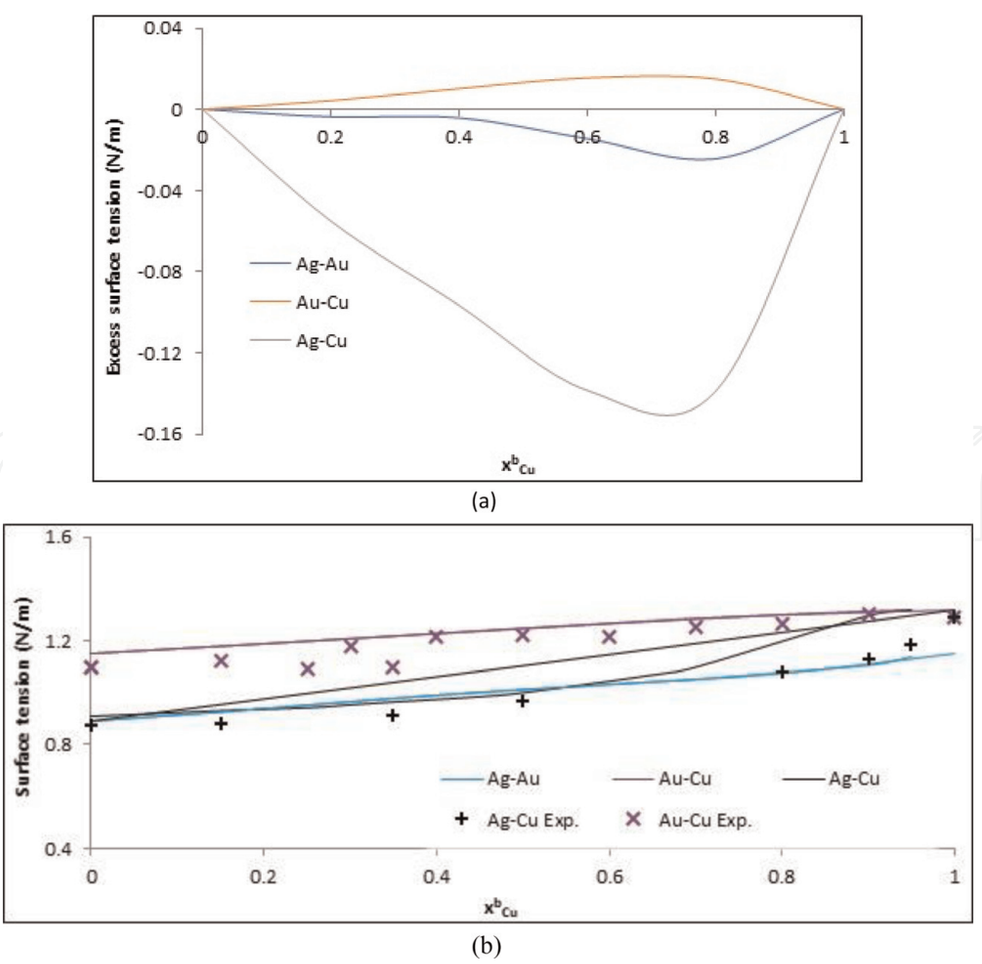


Figure 1.
(a) The excess binary surface tensions σ^{E} of three sub-binary systems in the Ag-Au-Cu ternary alloy at 1381 K and (b) the binary surface tensions σ of three sub-binary systems in the Ag-Au-Cu ternary alloy at 1381 K.

V_{Au} (m^3/mol) = 11.3 (1 + 0.8/10,000 (T/K − 1337.33))/1,000,000	[185]
V_{Ag} (m^3/mol) = 11.6 (1 + 0.98/10,000 (T/K − 1234.93))/1,000,000	[46]
V_{Cu} (m^3/mol) = 7.94 (1 + 1/10,000 (T/K − 1357.77))/1,000,000	[46]

Table 2.
Molar volume data for the Ag-Au-Cu alloy system.

System	A_{ij^0}	A_{ij^1}	A_{ij^2}
Ag-,Au	−16,402 + 1.14 T [198]	—	—
Ag-,Cu	17384.37 – 4.46438 T [199]	1660.74 – 2.31516 T [199]	—
Au-,Cu	−27,900 – T [200]	4730 [200]	3500 + 3.5 T [200]
Ag-,Au-,Cu	10,000 [199]	105,000 + 30 T [199]	−1000 [199]

Table 3.
Interaction parameters for excess Gibbs energy in the Ag-Au-Cu system.

liquid Ag, Bi, Sn, and the molar volumes (**Table 2**) [46, 185], were combined with the Gibbs energies (in order to calculate these energies, Redlich-Kister parameters given in **Table 3** are used) of the binary and ternary systems in order to calculate the surface tension of the liquid Ag-Au-Cu alloy systems using the Butler model.

5. Results and discussion

Geometrical models have been recently successfully applied to predict the surface tension of the Ni_3S_2-FeS-Cu_2S system, and the obtained results were in good agreement with those of experimental values [201]. Moreover, the surface tensions of the Sn-Ga-In ternary alloy systems were calculated from the surface tensions of the Sn-Ga, Ga-In and In-Sn sub-binary systems at 773 K using geometric models, such as the Kohler, Toop, and GSM, by some researchers [140]. In the Toop model, which is a classic asymmetric geometric model, it is very important to determine the asymmetric component [202]. As is expressed in previous papers [176–183, 199], if the deviations of the binary systems A-B and A-C from the ideal solution are similar, but differ markedly from those of the binary system B-C, then the A-B-C ternary system is asymmetric, and the ternary system mentioned above becomes symmetric. It can be readily said that the common component A in two sub-binary systems with thermodynamic similarities ought to be chosen as the asymmetric component in the asymmetric system in the Toop model. When the excess binary surface tensions σ^E of three sub-binary systems in the Ag-Au-Cu ternary alloy at 1381 K are compared, it can clearly be seen that the asymmetric component is Au because the two binary Ag-Au and Au-Cu systems are much more similar thermodynamically, as shown in **Figure 1**. The similarity coefficients for the calculated excess surface tension of the Ag-Au, Ag-Cu, and Au-Cu sub-binary alloys mentioned in GSM have been calculated and found as 0.934915, 0.600985, and 0.044179 (**Table 4**), respectively. On glancing at the values of the similarity coefficients, it is seen that none of the three similarity coefficients is approximately equivalent to unity. So, the asymmetric component in the ternary alloys is not easy to find. Here, $\xi_{Ag-Au} = 0.82$ and this means that Ag is relatively similar to Cu. It also indicates that someone has no alternative option but to

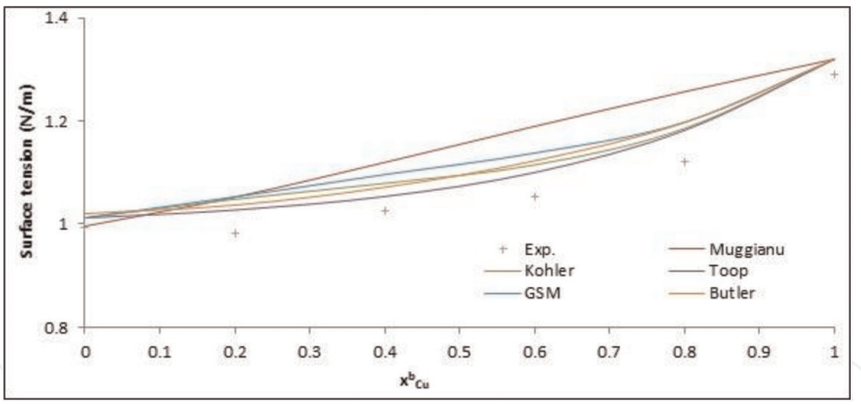
Deviation of sum of squares	η_1 = **8051.52**		η_2 = **560.51**		η_3 = **12126.95**	
Binary systems	Ag-Au	Ag-Cu	Au-Ag	Au-Cu	Cu-Ag	Cu-Au
Similarity coefficients	$\xi_{Ag\text{-}Au}$ = 0.934915		$\xi_{Au\text{-}Cu}$ = 0.044179		$\xi_{Cu\text{-}Ag}$ = 0.600985	

Table 4.
Deviation of sum of squares and similarity coefficients associated with surface tension in Ag-Au-Cu ternary system for GSM at 1381 K.

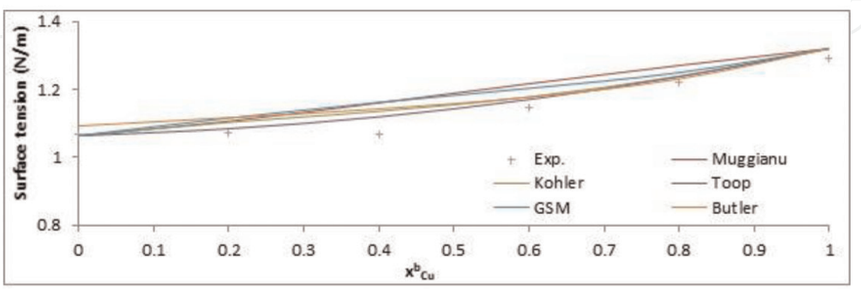
select Au as the asymmetric component. By investigating the abovementioned analysis, it can be concluded that the Ag-Au-Cu ternary system is not equivalent to the Muggianu, the Kohler, or the Toop models, so both the Muggianu model and the Kohler model cannot obtain the predicted values accurately. Therefore, the predicted surface tensions calculated by the GSM model can be recommended. In addition, according to the abovementioned analysis, one can conclude that the Ag-Au-Cu ternary system is exactly not determined by the symmetric models. A comparison of the surface tension values, which are calculated by the selected models, with the experimental values measured in literature [166, 167] for cross sections (**Figure 4**) (a) $z = x_{Ag}/x_{Au}$ = 1/3, (b) 1/1, (c) 3/1, (d) 2/5, and (e) 5/2, in the Ag-Au-Cu ternary system is shown in **Figure 2**. In addition, it is inferred from the discussion of the standard errors (**Table 5**) of the calculation results performed in the present study that the best model is the Toop model for all cross sections $z = x_{Ag}/x_{Au}$ = 1/3, 1/1, 3/1, 2/5, and 5/2, in the whole experimental range. Therefore, the excess surface tension curves of the Ag-Au, Ag-Cu, and Au-Cu sub-binaries calculated by Eq. (5) and Ag-Au-Cu ternary alloy systems at 1381 K by the Toop model are plotted in **Figure 3**, since the Toop model is found as the best model compared to the other models. For example, the comparison between the calculated surface tension for cross section x_{Ag}/x_{Au} = 1/3 in the Ag-Au-Cu alloy system and its experimental values at 1381 K is provided in **Table 6**.

On the other hand, a comparison of the surface tension values calculated via the Toop model with the experimental values measured in literature [167] for cross section $z = x_{Ag}/x_{Au}$ = 1/3, 1/1, 3/1, 2/5, and 5/2 in the Ag-Au-Cu ternary systemis given in **Figure 3a**. The Toop model is able to describe the experimental data for all ratios mentioned above. In addition to this, the dependency of the surface molar fraction of copper from bulk composition in the Ag-Au-Cu ternary system is calculated by the Butler equation and its plot is given in **Figure 3b**. One can obtain the distribution between interface region and bulk phase for all five components in Ag-Au-Cu ternary mixture calculating the surface mole fraction $x_{i,s}$ for a given bulk-phase mole fraction *xi*, B via the Butler equations (Eq. (18)). When the ratio of x_i,s/*xi*,b is equal to one, it yields no surface active enrichment/depletion owing to a reference line. At constant ratios $z = x_{Ag}/x_{Au}$ = 1/3, 1/1, 3/1, 2/5, and 5/2, the relative enrichment of all three components are depicted in **Figure 3b**. It can be seen at the relative high or low concentration x_{Ag}/x_{Au}, in the alloys mentioned above, that concentration of Cu can be found in the bulk phase and show no tendency to enter the surface region. It can be also seen from **Figure 3a** that the most striking aspects of the surface tension calculations carried out in the present study for all models can improve the surface tension values when Cu is added to the alloys with Ag and Cu contents.

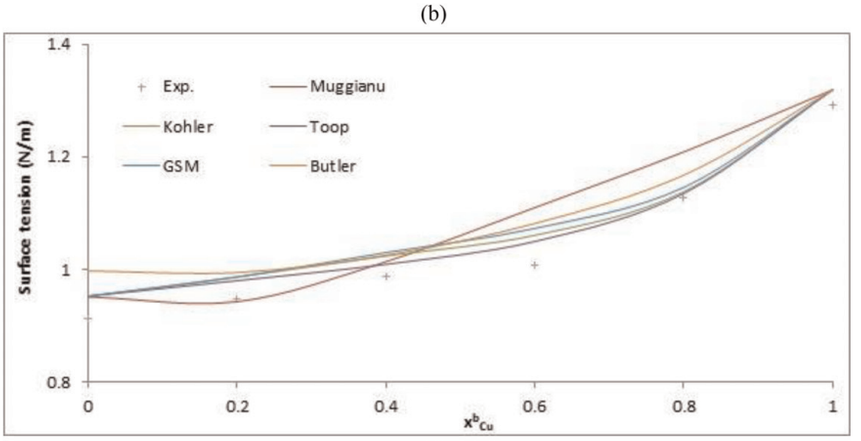
A comparison of the surface tension values calculated for binary Ag-Au, Ag-Cu, and Au-Cu alloys with the experimental values measured in literature except for Ag-Au [167] is given in **Figure 1b**. On the other hand, it is calculated by Calvo [169] that the surface tensions of Ag-Au binary alloys are found between 550 and 700 mN/m at a temperature of 1500 K. These results are also in agreement with those obtained experimentally by ref. [57], and those calculated in this study.



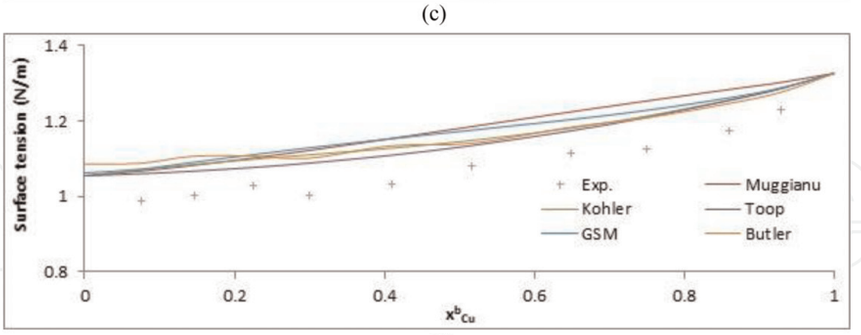
(a)



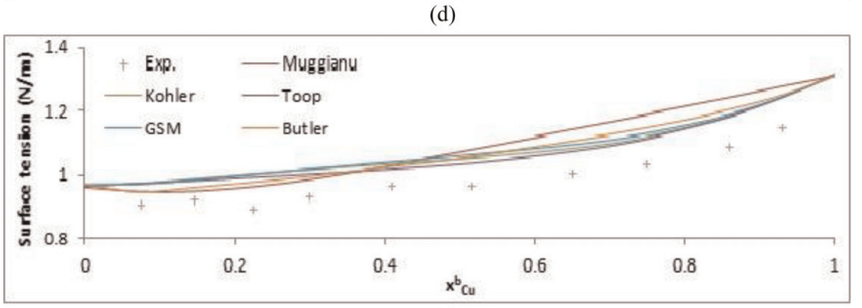
(b)



(c)



(d)

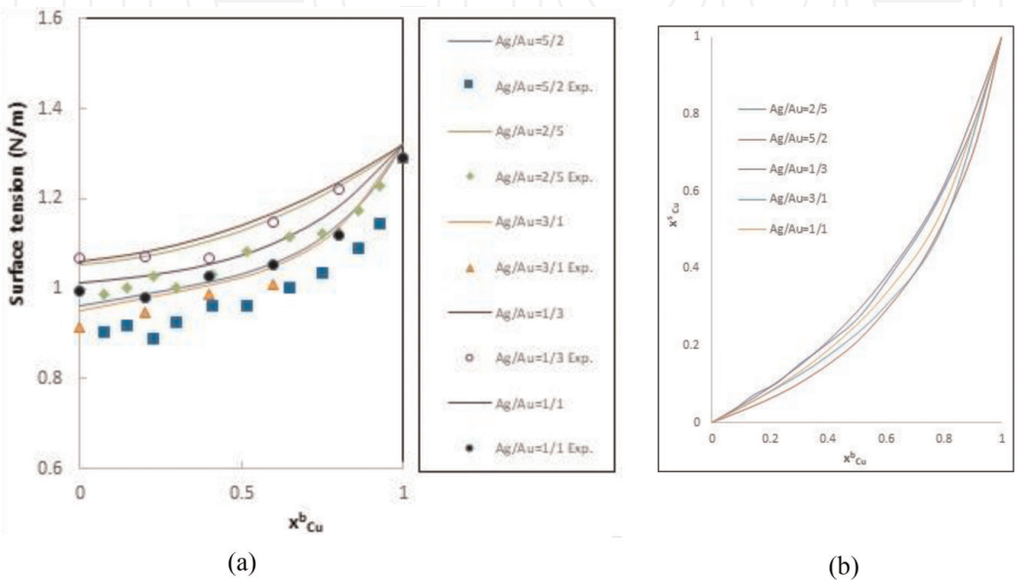


(e)

Figure 2.
A comparison of the surface tension values calculated by all the selected models with the experimental values measured in literature [166, 167] for cross section (a) z = x_{Ag}/x_{Au} = 1/3, (b) 1/1, (c) 3/1, (d) 2/5 and (e) 5/2, in the Ag-Au-Cu ternary system.

Sections	Muggianu	Kohler	Toop	GSM	Butler
Ag/Au = 1/1	0.038259	0.021469	0.016782	0.026046	0.021911
Ag/Au = 1/3	0.022055	0.014311	0.010802	0.020502	0.016629
Ag/Au = 3/1	0.023755	0.015022	0.012579	0.016889	0.022797
Ag/Au = 2/5	0.029510	0.023499	0.019233	0.028573	0.024484
Ag/Au = 5/2	0.028877	0.024461	0.021560	0.026221	0.024006

Table 5.
The standard errors associated with surface tension in Ag-Au-Cu ternary system.



(a) (b)

Figure 3.
(a) A comparison of the surface tension values calculated by Toop model with the experimental values measured in literature [166, 167] for all cross sections ($z = x_{Ag}/x_{Au}$ = 1/3, 1/1, 3/1, 2/5, and 5/2) in this figure concerning the Ag-Au-Cu ternary system. (b) Dependency of the surface-bulk composition of copper in the Ag-Au-Cu ternary system for all cross sections ($z = x_{Ag}/x_{Au}$ = 1/3, 1/1, 3/1, 2/5, and 5/2).

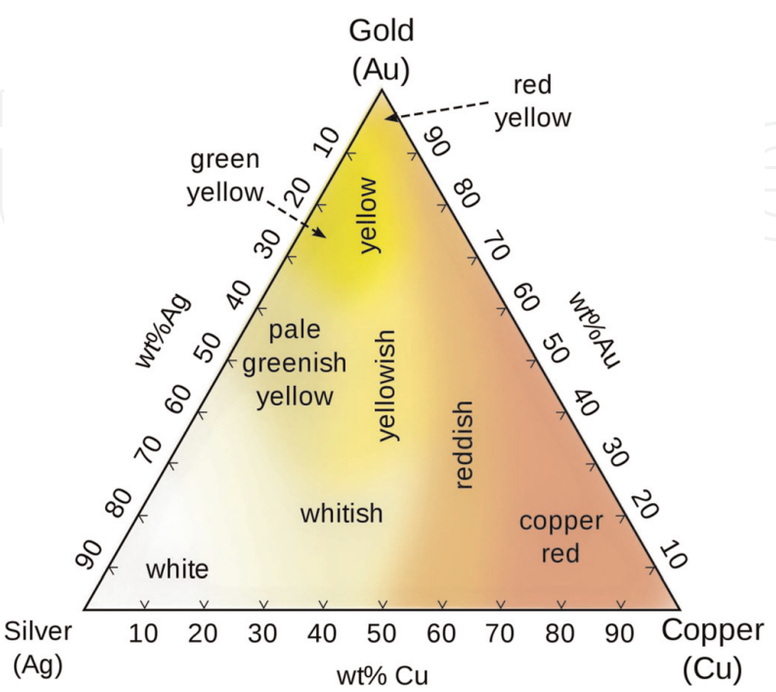


Figure 4.
A phase diagram of the Ag-Au-Cu ternary alloy [203].

x^b_{Cu}	Muggianu	Kohler	Toop	GSM	Butler	Exp.
0	1.0620	1.0620	1.0620	1.0620	1.0922	1.067
0.2	1.1051	1.1018	1.0828	1.1079	1.1163	1.071
0.4	1.1593	1.1346	1.1181	1.1474	1.1423	1.069
0.6	1.2160	1.1770	1.1692	1.1893	1.1767	1.147
0.8	1.2701	1.2382	1.2365	1.2433	1.2307	1.220
1	1.3200	1.3200	1.3200	1.3200	1.3200	1.291

Table 6.
A comparison between the calculated surface tension for cross section x_{Ag}/x_{Au} = 1/3 in the Ag-Au-Cu alloy system and its experimental values at 1381 K [167].

Taking into consideration the fact that there is no sufficient experimental thermophysical data in literature, it is envisaged that the results obtained from the theoretical analysis of the alloy systems in the present work might fill the research gap on the thermophysical properties such as surface tension. Therefore, there are no agreements among the models except for the cross sections $z = x_{Ag}/x_{Au}$ = 1/1, 3/1. According to the researchers, the high temperature might be one of the factors that mainly affect the surface tension so that the oxidation at high temperatures directly affects surface tension and will give rise to decreased values.

The major experimental measuring difficulty in surface tension measurements of the multicomponent alloys at high temperatures lies in the control of the processing atmosphere existing in device medium. Taking into account the difficulty in measuring the surface tension of multicomponent alloys, surface tension of the multicomponent alloys was calculated by calculating the surface tension of two-component liquid alloys and demonstrated their general applicability in this study.

6. Conclusions

For cross sections $z = x_{Ag}/x_{Au}$ = 1/3, 1/1, 3/1, 2/5, and 5/2, the surface tension values of the treated alloys Ag-Au-Cu have been modeled and analyzed theoretically by using such approaches to geometric models, as the Muggianu, the Kohler, the Toop, and the GSM models, and Butler's equation. Considering that the predictions of all models are generally consistent with the experimental findings, some conclusions are given as follows: the surface tension values increase as the composition of Cu increases. It is seen at the relative high or low concentration x_{Ag}/x_{Au} in the above-mentioned alloys that concentration of Cu can be found in the bulk phase and shows no tendency to enter the surface region. It is inferred from the calculations performed for all models that the Toop method yields in a great promise to efficiently determine the physical properties such as surface tension data of alloys Ag-Au-Cu.

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References

[1] Keene B. Review of data for the surface tension of pure metals. International Materials Reviews. 1993; **38**:157-192

[2] Gale W, Totemeier T, editors. Smithells Metals Reference Book. 8th ed. Amsterdam, Amsterdam Boston, Heidelberg, London, New York, Oxford, Paris, San Diego, San Francisco, Singapore, Sydney, Tokyo: Elsevier; 2004

[3] Man KF. Surface tension measurements of liquid metals by the quasi-containerless pendant drop method. International Journal of Thermophysics. 2000;**21**:793-804

[4] Egry I, Lohoefer G, Jacobs G. Surface tension of liquid metals: Results from measurements on ground and in space. Physical Review Letters. 1995;**75**:4043

[5] Egry I, Sauerland S. Containerless processing of undercooled melts: Measurements of surface tension and viscosity. Materials Science and Engineering A. 1994;**178**:73-76

[6] Egry I, Brillo Jr. Surface tension and density of liquid metallic alloys measured by electromagnetic levitation. Journal of Chemical & Engineering Data. 2009;**54**:2347-2352

[7] Lee J, Kiyose A, Nakatsuka S, Nakamoto M, Tanaka T. Improvements in surface tension measurements of liquid metals having low capillary constants by the constrained drop method. ISIJ International. 2004;**44**:1793-1799

[8] Ricci E, Giuranno D, Sobczak N. Further development of testing procedures for high temperature surface tension measurements. Journal of Materials Engineering and Performance. 2013;**22**:3381-3388

[9] Falke W, Schwaneke A, Nash R. Surface tension of zinc: The positive temperature coefficient. Metallurgical Transactions B. 1977;**8**:301-303

[10] Brillo J. Thermophysical Properties of Multicomponent Liquid Alloys. Berlin, Boston: Walter de Gruyter GmbH & Co KG; 2016

[11] Amore S, Giuranno D, Novakovic R, Ricci E, Nowak R, Sobczak N. Thermodynamic and surface properties of liquid Ge-Si alloys. Calphad. 2014;**44**: 95-101

[12] Thiessen D, Man K. Surface tension measurement. In: The Measurement, Instrumentation and Sensors Handbook on CD-ROM. Boca Raton: CRC Press; 1999

[13] Defay R, Hommelen JR. I. Measurement of dynamic surface tensions of aqueous solutions by the oscillating jet method. Journal of Colloid Science. 1958;**13**:553-564

[14] Defay R, Pétré G. Correcting surface tension data obtained by the oscillating jet method. Journal of Colloid Science. 1962;**17**:565-569

[15] Bellizia G, Megaridis CM, McNallan M, Wallace DB. A capillary-jet instability method for measuring dynamic surface tension of liquid metals. In: Proceedings of the Royal Society of London A: Mathematical, Physical and Engineering Sciences. The Royal Society; 2003. pp. 2195-2214

[16] Dubberstein T, Heller H-P, Klostermann J, Schwarze R, Brillo J. Surface tension and density data for Fe-Cr-Mo, Fe-Cr-Ni, and Fe-Cr-Mn-Ni steels. Journal of Materials Science. 2015;**50**:7227-7237

[17] Howell E, Megaridis C, McNallan M. Dynamic surface tension measurements of molten Sn/Pb solder using oscillating slender elliptical jets. International

Journal of Heat and Fluid Flow. 2004;**25**: 91-102

[18] Gancarz T, Moser Z, Gąsior W, Pstruś J, Henein H. A comparison of surface tension, viscosity, and density of Sn and Sn-Ag alloys using different measurement techniques. International Journal of Thermophysics. 2011;**32**: 1210-1233

[19] Roach S, Henein H. A dynamic approach to determining the surface tension of a fluid. Canadian Metallurgical Quarterly. 2003;**42**: 175-186

[20] Roach SJ, Henein H. A new method to dynamically measure the surface tension, viscosity, and density of melts. Metallurgical and Materials Transactions B. 2005;**36**:667-676

[21] Stückrad B, Hiller W, Kowalewski T. Measurement of dynamic surface tension by the oscillating droplet method. Experiments in Fluids. 1993;**15**: 332-340

[22] Cao C, Zhang L, Bai X, Duan L, Wang F. Measurement of surface tension and specific heat of Ni-18.8 at.% Si alloy melt by containerless processing. Journal of Materials Science. 2011;**46**:6243-6247

[23] Schmitz J, Brillo J, Egry I, Schmid-Fetzer R. Surface tension of liquid Al-Cu binary alloys. International Journal of Materials Research. 2009;**100**:1529-1535

[24] Dai F, Cao C, Wei B. Thermophysical properties of Ni-5% Sn alloy melt. Science in China Series G: Physics Mechanics and Astronomy. 2006;**49**:236-245

[25] Matsumoto T, Fujii H, Ueda T, Kamai M, Nogi K. Measurement of surface tension of molten copper using the free-fall oscillating drop method. Measurement Science and Technology. 2005;**16**:432

[26] Wang H, Cao C, Wei B. Thermophysical properties of a highly superheated and undercooled Ni-Si alloy melt. Applied Physics Letters. 2004;**84**: 4062-4064

[27] Egry I, Giffard H, Schneider S. The oscillating drop technique revisited. Measurement Science and Technology. 2005;**16**:426

[28] Kobatake H, Brillo J, Schmitz J, Pichon P-Y. Surface tension of binary Al-Si liquid alloys. Journal of Materials Science. 2015;**50**:3351-3360

[29] Ravera F, Loglio G, Kovalchuk VI. Interfacial dilational rheology by oscillating bubble/drop methods. Current Opinion in Colloid & Interface Science. 2010;**15**:217-228

[30] Seyhan I, Egry I. The surface tension of undercooled binary iron and nickel alloys and the effect of oxygen on the surface tension of Fe and Ni. International Journal of Thermophysics. 1999;**20**:1017-1028

[31] Schneider S, Egry I, Seyhan I. Measurement of the surface tension of undercooled liquid $Ti_{90}Al_6V_4$ by the oscillating drop technique. International Journal of Thermophysics. 2002;**23**: 1241-1248

[32] Fujii H, Matsumoto T, Izutani S, Kiguchi S, Nogi K. Surface tension of molten silicon measured by microgravity oscillating drop method and improved sessile drop method. Acta Materialia. 2006;**54**:1221-1225

[33] Brillo J, Plevachuk Y, Egry I. Surface tension of liquid Al-Cu-Ag ternary alloys. Journal of Materials Science. 2010;**45**:5150-5157

[34] Egry I, Brillo J, Holland-Moritz D, Plevachuk Y. The surface tension of liquid aluminium-based alloys. Materials Science and Engineering A. 2008;**495**:14-18

[35] Schick M, Brillo J, Egry I. Thermophysical properties of liquid Co-Cu-Ni alloys. International Journal of Cast Metals Research. 2009;**22**:82-85

[36] Han X, Wei B. Thermophysical properties of undercooled liquid Co-Mo alloys. Philosophical Magazine. 2003;**83**: 1511-1532

[37] Brillo J, Egry I. Density and surface tension of electromagnetically levitated Cu-Co-Fe alloys. International Journal of Thermophysics. 2007;**28**:1004-1016

[38] Wang H, Luo B, Qin T, Chang J, Wei B. Surface tension of liquid ternary Fe-Cu-Mo alloys measured by electromagnetic levitation oscillating drop method. The Journal of Chemical Physics. 2008;**129**:124706

[39] Brillo J, Egry I. Surface tension of nickel, copper, iron and their binary alloys. Journal of Materials Science. 2005;**40**:2213-2216

[40] Chang J, Wang H, Zhou K, Wei B. Surface tension measurement of undercooled liquid Ni-based multicomponent alloys. Philosophical Magazine Letters. 2012;**92**:428-435

[41] Henein H. The transition from free stream flow to dripping in draining vessels. Canadian Metallurgical Quarterly. 2005;**44**:261-264

[42] Trybula M, Gancarz T, Gasior W, Pasturel A. Bulk and surface properties of liquid Al-Li and Li-Zn alloys. Metallurgical and Materials Transactions A. 2014;**45**:5517-5530

[43] Tate T. XXX. On the magnitude of a drop of liquid formed under different circumstances. The London, Edinburgh, and Dublin Philosophical Magazine and Journal of Science. 1864;**27**:176-180

[44] Harkins WD, Brown F. The determination of surface tension (free surface energy), and the weight of falling drops: The surface tension of water and benzene by the capillary height method. Journal of the American Chemical Society. 1919;**41**:499-524

[45] Park SH, Um YS, Hur BY. Rheological properties of molten Al-Cu alloys for manufacturing metallic foam. Solid State Phenomena. 2006;**116**: 656-660

[46] Lee J, Shimoda W, Tanaka T. Surface tension and its temperature coefficient of liquid Sn-X (X= Ag, Cu) alloys. Materials Transactions. 2004;**45**: 2864-2870

[47] Plevachuk Y, Sklyarchuk V, Gerbeth G, Eckert S, Novakovic R. Surface tension and density of liquid Bi-Pb, Bi-Sn and Bi-Pb-Sn eutectic alloys. Surface Science. 2011;**605**:1034-1042

[48] Chang C-H, Coltharp KA, Park SY, Franses EI. Surface tension measurements with the pulsating bubble method. Colloids and Surfaces A: Physicochemical and Engineering Aspects. 1996;**114**:185-197

[49] Arashiro EY, Demarquette NR. Use of the pendant drop method to measure interfacial tension between molten polymers. Materials Research. 1999;**2**: 23-32

[50] Kumar A, Lehnert-Batar S, Mäder R, Windhab EJ. Measurement of the surface and interfacial tension from maximum volume of a pendant drop. Journal of Colloid and Interface Science. 2001;**244**:113-122

[51] Semmler A, Kohler H-H. Surface properties of alkylpyridinium chlorides and the applicability of the pendant drop technique. Journal of Colloid and Interface Science. 1999;**218**:137-144

[52] Andreas J, Hauser E, Tucker W. Boundary tension by pendant drops 1. The Journal of Physical Chemistry. 1938; **42**:1001-1019

[53] Stauffer CE. The measurement of surface tension by the pendant drop technique. The Journal of Physical Chemistry. 1965;**69**:1933-1938

[54] Bashforth F. In: Bashforth F, Adams JC, editors. An Attempt to Test the Theories of Capillary Action by Comparing the Theoretical and Measured Forms of Drops of Fluid, with an Explanation of the Method of Integration Employed in Constructing the Tables which Give the Theoretical Forms of Such Drops. UK: Cambridge University Press; 1883

[55] Quincke G. Ueber die Capillaritätsconstanten des Quecksilbers. Annalen der Physik. 1858; **181**:1-48

[56] Padday JF. The profiles of axially symmetric menisci. Philosophical Transactions of the Royal Society of London A: Mathematical Physical and Engineering Sciences. 1971;**269**:265-293

[57] Bernard G, Lupis C. The surface tension of liquid silver alloys: Part I. Silver-gold alloys. Metallurgical Transactions. 1971;**2**:555-559

[58] Bachmann J, Ellies A, Hartge K. Development and application of a new sessile drop contact angle method to assess soil water repellency. Journal of Hydrology. 2000;**231**:66-75

[59] Siwiec G, Oleksiak B, Smalcerz A, Wieczorek J. Surface tension of Cu-Ag alloys/Napiecie Powierzchniowe Stopów Cu-Ag. Archives of Metallurgy and Materials. 2013;**58**:193-195

[60] Tanaka T, Matsuda M, Nakao K, Katayama Y, Kaneko D, Hara S, et al. Measurement of surface tension of liquid Ga-base alloys by a sessile drop method. Zeitschrift fuer MetaIlkunde. 2001;**92**:1242-1246

[61] Chao KL, Lian SS, Wu CY, Chang KL. Surface tension measurement of liquid Fe-Si-B alloy by sessile drop method. Key Engineering Materials. 2014;**573**:9-12

[62] Egry I, Ricci E, Novakovic R, Ozawa S. Surface tension of liquid metals and alloys—Recent developments. Advances in Colloid and Interface Science. 2010; **159**:198-212

[63] Krasovskyy V, Naidich Y. Surface tension and specific volume of copper-titanium melts measured by the sessile drop method. Journal of Adhesion Science and Technology. 2004;**18**: 465-471

[64] Feng X, Liu L-X, Yang R-H, Zhao H-K, Liang F, Zhang C. Surface tension of molten Ni-(Cr, Co, W) alloys and segregation of elements. Transactions of Nonferrous Metals Society of China. 2008;**18**:1184-1188

[65] Guo Z, Hindler M, Yuan W, Mikula A. Density and surface tension of liquid Bi-Cu-Sn alloys. Monatshefte für Chemie-Chemical Monthly. 2012;**143**:1617-1622

[66] Guo Z, Hindler M, Yuan W, Mikula A. The density and surface tension of In-Sn and Cu-In-Sn alloys. Monatshefte für Chemie-Chemical Monthly. 2011;**142**: 579-584

[67] Giuranno D, Tuissi A, Novakovic R, Ricci E. Surface tension and density of Al-Ni alloys. Journal of Chemical & Engineering Data. 2010;**55**:3024-3028

[68] Plevachuk Y, Sklyarchuk V, Eckert S, Gerbeth G, Novakovic R. Thermophysical properties of the liquid Ga-In-Sn eutectic alloy. Journal of Chemical & Engineering Data. 2014;**59**: 757-763

[69] Guo Z, Li S, Mikula A, Yuan W. Surface tension of liquid Au-Bi-Sn alloys. Rare Metals. 2012;**31**:250-254

[70] Oleksiak B, Łabaj J, Wieczorek J, Blacha-Grzechnik A, Burdzik R. Surface

tension of Cu-Bi alloys and wettability in a liquid alloy-refractory material-gaseous phase system. Archives of Metallurgy and Materials. 2014;**59**: 282-286

[71] Kucharski M, Fima P. The surface tension and density of liquid Ag-Bi, Ag-Sn, and Bi-Sn alloys. Monatshefte für Chemie/Chemical Monthly. 2005;**136**: 1841-1846

[72] Lee JH, Lee DN. Use of thermodynamic data to calculate surface tension and viscosity of Sn-based soldering alloy systems. Journal of Electronic Materials. 2001;**30**:1112

[73] Pstruś J, Moser Z, Gąsior W. Surface properties of liquid In-Zn alloys. Applied Surface Science. 2011;**257**: 3867-3871

[74] Sklyarchuk V, Plevachuka Y, Kaban I, Novakovic R. Surface properties and wetting behavior of liquid Ag-Sb-Sn alloys. Journal of Mining and Metallurgy, Section B: Metallurgy. 2012; **48**:443-448

[75] Sharan A, Nagasaka T, Cramb A. Surface tensions of liquid Fe-Cr and Fe-Cr-N alloys. Metallurgical and Materials Transactions B. 1994;**25**:626-628

[76] Willner J, Siwiec G, Botor J. The surface tension of liquid Cu-Fe-Sb alloys. Applied Surface Science. 2010; **256**:2939-2943

[77] Fima P. Surface tension and density of liquid Sn-Ag alloys. Applied Surface Science. 2011;**257**:3265-3268

[78] Ohira C, Fujii H, Morisada Y. Temperature dependence of surface tension of Sn-Ag alloys. Journal of Materials Engineering and Performance. 2014;**23**:1561-1567

[79] Mishchuk N, Fainerman V, Kovalchuk V, Miller R, Dukhin S. Studies of concentrated surfactant solutions using the maximum bubble pressure method. Colloids and Surfaces A: Physicochemical and Engineering Aspects. 2000;**175**:207-216

[80] Drelich J, Fang C, White C. Measurement of interfacial tension in fluid-fluid systems. Encyclopedia of Surface and Colloid Science. 2002;**3**: 3158-3163

[81] Mishchuk NA, Fainerman VB, Kovalchuk VI, Miller R, Duhkin SS. Studies of concentrated surfactant solutions using the maximum bubble pressure method. Colloid Surface A. 2000;**175**:207-216

[82] Sugden S. XCVII—The determination of surface tension from the maximum pressure in bubbles. Journal of the Chemical Society, Transactions. 1922;**121**:858-866

[83] Rehbinder P. Dependence of surface activity and surface tension of solutions upon temperature and concentration. Zeitschrift für Physikalische Chemie. 1924;**111**:447-464

[84] Gasior W, Pstruś J, Moser Z, Krzyżak A, Fitzner K. Surface tension and thermodynamic properties of liquid Ag-Bi solutions. Journal of Phase Equilibria. 2003;**24**:40-49

[85] Gasior W, Moser Z, Pstruś J. Surface tension, density, and molar volume of liquid Sb-Sn alloys: Experiment versus modeling. Journal of Phase Equilibria. 2003;**24**:504-510

[86] Moser Z, Gasior W, Pstruś J. Surface tension of liquid Ag-Sn alloys: Experiment versus modeling. Journal of Phase Equilibria. 2001;**22**:254-258

[87] Liu X, Inohana Y, Takaku Y, Ohnuma I, Kainuma R, Ishida K, et al. Experimental determination and thermodynamic calculation of the phase equilibria and surface tension in the

Sn-Ag-In system. Journal of Electronic Materials. 2002;**31**:1139-1151

[88] Pstruś J, Moser Z, Gąsior W, Dębski A. Surface tension and density measurements of liquid Sn-Zn alloys. Experiment vs. SURDATdatabase of Pb-free solders. Archives of Metallurgy and Materials. 2006;**51**:335-343

[89] Moser Z, Gasior W, Bukat K, Pstrus J, Kisiel R, Sitek J, et al. Pb-free solders: Part III. Wettability testing of Sn-Ag-Cu-Bi alloys with Sb additions. Journal of Phase Equilibria and Diffusion. 2007;**28**:433-438

[90] Kraut J, Stern W. The density of gold-silver-copper alloys and its calculation from the chemical composition. Gold Bulletin. 2000;**33**: 52-55

[91] Moser Z, Gasior W, Pstrus J, Ishihara S, Liu XJ, Ohnuma I, et al. Surface tension and density measurements of Sn-Ag-Sb liquid alloys and phase diagram calculations of the Sn-Ag-Sb ternary system. Materials Transactions. 2004;**45**:652-660

[92] Gąsior W, Moser Z, Pstruś J. Measurements of the surface tension and density of TIN based Sn-Ag-Cu-Sb liquid alloys. Archives of Metallurgy and Materials. 2004;**49**:155-167

[93] Moser Z, Gasior W, Pstruś J, Ksiezarek S. Surface-tension measurements of the eutectic alloy (Ag-Sn 96.2 at.%) with Cu additions. Journal of Electronic Materials. 2002;**31**: 1225-1229

[94] Moser Z, Gasior W, Pstrus J. Surface tension measurements of the Bi-Sn and Sn-Bi-Ag liquid alloys. Journal of Electronic Materials. 2001;**30**:1104-1111

[95] Moser Z, Gąsior W, Bukat K, Pstruś J, Kisiel R, Sitek J, et al. Pb-free solders: Part 1. Wettability testing of Sn-Ag-Cu alloys with Bi additions. Journal of Phase Equilibria and Diffusion. 2006;**27**: 133-139

[96] Moser Z, Gasior W, Ishida K, Ohnuma I, Bukat K, Pstrus J, et al. Experimental wettability studies combined with the related properties from data bases for lead-free soldering materials. In: CALPHAD XXXIII Program & Abstract; 2004

[97] Moser Z, Gąsior W, Pstruś J, Ohnuma I, Ishida K. Influence of Sb additions on surface tension and density of Sn-Sb, Sn-Ag-Sb and Sn-Ag-Cu-Sb alloys: Experiment vs. modeling. Zeitschrift fuer MetaIlkunde. 2006;**97**: 365-370

[98] Gasior W, Moser Z, Pstruś J, Bukat K, Sitek J, Kisiel R. $(Sn\text{-}Ag)_{eut}$+Cu soldering materials, part I: Wettability studies. Journal of Phase Equilibria and Diffusion. 2004;**25**:115-121

[99] Moser Z, Gasior W, Pstrus J, Ksiezarek S. Surface tension and density of the (Ag-Sn) eut+Cu liquid alloys. Journal of Electronic Materials. 2002;**31**: 1225-1229

[100] Moser Z, Gąsior W, Ishida K, Ohnuma I, Liu X, Bukat K, et al. Experimental wettability studies combined with the related properties from data base for tin based alloys with silver, copper, bismuth and antimony additions. In: 134th Annual Meeting & Exhibition, Book of Final Program; 2005. pp. 13-17

[101] Moser Z, Gasior W, Debski A. Database of Pb-free soldering materials, surface tension and density, experiment vs. modeling. Data Science Journal. 2005;**4**:195-208

[102] Gasior W, Moser Z, Bukat K, Pstrus J, Sitek J, Kisiel R. Experimental wettability studies of tin based liquid alloys with Ag, Cu, Bi and Sb additions. TOFA; 2004

[103] Jacob KT, Jayadevan KP. $(Sn-Ag)_{eut}$+Cu soldering materials, part II: Electrical and mechanical studies. Journal of Phase Equilibria and Diffusion. 2004;**25**:122-124

[104] Gasior W, Pstrus J, Moser Z. Influence of zinc on the surface tension, density and molar volume of (Ag-Sn) (EUT)+ Zn liquid alloys. Archives of Metallurgy and Materials. 2016;**61**: 361-367

[105] Goicoechea J, Garcia-Cordovilla C, Louis E, Pamies A. Surface tension of binary and ternary aluminium alloys of the systems Al-Si-Mg and Al-Zn-Mg. Journal of Materials Science. 1992;**27**: 5247-5252

[106] Pstruś J. Surface tension and density of liquid In-Sn-Zn alloys. Applied Surface Science. 2013;**265**:50-59

[107] Quested P, Brooks R, Chapman L, Morrell R, Youssef Y, Mills K. Measurement and estimation of thermophysical properties of nickel based superalloys. Materials Science and Technology. 2009;**25**:154-162

[108] Mukai K, Matsushita T, Mills KC, Seetharaman S, Furuzono T. Surface tension of liquid alloys—A thermodynamic approach. Metallurgical and Materials Transactions B. 2008;**39**: 561-569

[109] Mills K, Su Y. Review of surface tension data for metallic elements and alloys: Part 1-pure metals. International Materials Reviews. 2006;**51**:329-351

[110] Mills KC, Youssef YM, Li Z, Su Y. Calculation of thermophysical properties of Ni-based superalloys. ISIJ International. 2006;**46**:623-632

[111] Aune R, Battezzati L, Brooks R, Egry I, Fecht H-J, Garandet J-P, et al. Surface tension and viscosity of industrial alloys from parabolic flight experiments—Results of the thermo lab project. Microgravity Science and Technology. 2005;**16**:11-14

[112] Aune R, Battezzati L, Brooks R, Egry I, Fecht H-J, Garandet J-P, et al. Measurement of thermophysical properties of liquid metallic alloys in a ground-and microgravity based research programme—The ThermoLab project. Microgravity Science and Technology. 2005;**16**:7-10

[113] Li Z, Mukai K, Zeze M, Mills K. Determination of the surface tension of liquid stainless steel. Journal of Materials Science. 2005;**40**:2191-2195

[114] Su Y, Li Z, Mills K. Equation to estimate the surface tensions of stainless steels. Journal of Materials Science. 2005;**40**:2201-2205

[115] Su Y, Mills K, Dinsdale A. A model to calculate surface tension of commercial alloys. Journal of Materials Science. 2005;**40**:2185-2190

[116] Mills KC, Su Y, Li Z, Brooks RF. Equations for the calculation of the thermo-physical properties of stainless steel. ISIJ International. 2004;**44**: 1661-1668

[117] Li Z, Mills KC, McLean M, Mukai K. Measurement of the density and surface tension of Ni-based superalloys in the liquid and mushy states. Metallurgical and Materials Transactions B. 2005;**36**:247-254

[118] Mills K, Brooks R. Measurements of thermophysical properties in high temperature melts. Materials Science and Engineering A. 1994;**178**:77-81

[119] Margrave JL. Determination of thermophysical properties of liquid metals at high temperatures by levitation methods. Materials Science and Engineering A. 1994;**178**:83-88

[120] Brooks R, Monaghan B, Barnicoat A, McCabe A, Mills K, Quested P. The

physical properties of alloys in the liquid and "mushy" states. International Journal of Thermophysics. 1996;**17**: 1151-1161

[121] Brooks R, Day A, Mills K, Quested P. Physical property measurements for the mathematical modeling of fluid flow in solidification processes. International Journal of Thermophysics. 1997;**18**: 471-480

[122] Keene B, Mills K, Kasama A, McLean A, Miller W. Comparison of surface tension measurements using the levitated droplet method. Metallurgical Transactions B. 1986;**17**:159-162

[123] Keene B, Mills K, Brooks R. Surface properties of liquid metals and their effects on weldability. Materials Science and Technology. 1985;**1**:559-567

[124] Keene BJ, Mills K, Bryant J, Hondros E. Effects of interaction between surface active elements on the surface tension of iron. Canadian Metallurgical Quarterly. 1982;**21**: 393-403

[125] Zeng BFG. Influencing the wettability of HSS-steel by addition of alloyingelmentsto the zink bath. In: Proceeding of EMC; 2009. pp. 1-18

[126] Adhikari D, Jha I, Singh B. Transport and surface properties of molten Al-Mn alloy. Advanced Materials Letters. 2012;**3**:226-230

[127] Brillo J, Lauletta G, Vaianella L, Arato E, Giuranno D, Novakovic R, et al. Surface tension of liquid Ag-Cu binary alloys. ISIJ International. 2014; **54**:2115-2119

[128] Mills KC. Recommended Values of Thermophysical Properties for Selected Commercial Alloys. UK: Cambridge University Press; 2002

[129] Ohnuma I, Ishida K, Moser Z, Gsior S, Bukat K, Pstru J, et al. Pb-free solders: Part II. Application of ADAMIS database in modeling of Sn-Ag-Cu alloys with Bi additions. Journal of Phase Equilibria and Diffusion. 2006;**27**:245

[130] Liu XJ, Yamaki T, Ohnuma I, Kainuma R, Ishida K. Thermodynamic calculations of phase equilibria, surface tension and viscosity in the In-Ag-X (X = Bi, Sb) system. Materials Transactions. 2004;**45**:637-645

[131] Hoffmann M, Wynblatt P. Surface composition of ternary Cu-Ag-Au alloys: Part I. Experimental results. Metallurgical Transactions A. 1991;**22**: 1833-1840

[132] Gancarz T. Physicochemical properties of Sb-Sn-Zn alloys. Journal of Electronic Materials. 2014;**43**:4374-4385

[133] Yadav S, Jha L, Jha I, Singh B, Koirala R, Adhikari D. Prediction of thermodynamic and surface properties of Pb-Hg liquid alloys at different temperatures. Philosophical Magazine. 2016;**96**:1909-1925

[134] Gancarz T, Pstru J, Gsior W, Henein H. Physicochemical properties of Sn-Zn and SAC+ Bi alloys. Journal of Electronic Materials. 2013;**42**:288

[135] Egry I. The surface tension of binary alloys: Simple models for complex phenomena. International Journal of Thermophysics. 2005;**26**: 931-939

[136] McNallan MJ, Debroy T. Effect of temperature and composition on surface tension in Fe-Ni-Cr alloys containing sulfur. Metallurgical Transactions B. 1991;**22**:557-560

[137] Novakovic R, Tanaka T. Bulk and surface properties of Al-Co and Co-Ni liquid alloys. Physica B: Condensed Matter. 2006;**371**:223-231

[138] Costa C, Delsante S, Borzone G, Zivkovic D, Novakovic R.

Thermodynamic and surface properties of liquid Co-Cr-Ni alloys. The Journal of Chemical Thermodynamics. 2014;**69**: 73-84

[139] Gąsior W, Fima P, Moser Z. Modeling of the thermodynamic properties of liquid Fe-Ni and Fe-Co alloys from the surface tension data. Archives of Metallurgy and Materials. 2011;**56**:13-23

[140] Yan L, Zheng S, Ding G, Xu G, Qiao Z. Surface tension calculation of the Sn-Ga-In ternary alloy. Calphad. 2007;**31**:112-119

[141] Trybula M, Gancarz T, Gąsior W. Density, surface tension and viscosity of liquid binary Al-Zn and ternary Al-Li-Zn alloys. Fluid Phase Equilibria. 2016; **421**:39-48

[142] Tanaka T. Prediction of phase diagrams in nano-sized binary alloys. Materials Science Forum. 2010;**653**:55-75

[143] Prasad L, Mikula A. Surface segregation and surface tension in Al-Sn-Zn liquid alloys. Physica B: Condensed Matter. 2006;**373**:142-149

[144] Novakovic R, Brillo J. Thermodynamics, thermophysical and structural properties of liquid Fe-Cr alloys. Journal of Molecular Liquids. 2014;**200**:153-159

[145] Prasad L, Xie Y, Mikula A. Lead free solder materials In-Sn-Zn system. Journal of Non-Crystalline Solids. 1999; **250**:316-320

[146] Terzieff P, Li Z, Knott S, Mikula A. Physicochemical properties of liquid Ag-Bi-Sn. Physica B: Condensed Matter. 2007;**388**:312-317

[147] Terzieff P. Surface tension, viscosity and concentration fluctuations in liquid Ag-In-Sn. Physics and Chemistry of Liquids. 2006;**44**:115-125

[148] Terzieff P. Concentration fluctuations and surface tension in liquid Au-Sn-Zn. Journal of Materials Science. 2005;**40**:3759-3763

[149] Wang X, Li W. Models to estimate viscosities of ternary metallic melts and their comparisons. Science in China, Series B: Chemistry. 2003;**46**:280-289

[150] Liu X-j, Wang C-p, Ohnuma I, Kainuma R, Ishida K. Development of thermodynamic and kinetic databases in micro-soldering alloy systems and their applications. Progress in Natural Science: Materials International. 2011;**21**:97-110

[151] Prasad L, Jha R. Correlation between bulk and surface properties of ternary Ag-Sn-Zn liquid alloys and concerned binaries. Physics and Chemistry of Liquids. 2007;**45**:149-167

[152] Kaban I, Gruner S, Hoyer W. Surface tension and density in liquid Ag-Cu-Sn alloys. Journal of Non-Crystalline Solids. 2007;**353**:3717-3721

[153] Kaban IG, Gruner S, Hoyer W. Experimental and theoretical study of the surface tension in liquid Ag-Cu-Sn alloys. Monatshefte für Chemie/ Chemical Monthly. 2005;**136**:1823-1828

[154] Akinlade O, Singh R. Bulk and surface properties of liquid In-Cu alloys. Journal of Alloys and Compounds. 2002; **333**:84-90

[155] Ohnuma I, Liu X, Ohtani H, Ishida K. Thermodynamic database for phase diagrams in micro-soldering alloys. Journal of Electronic Materials. 1999;**28**: 1164-1171

[156] Tanaka T, Hack K, Hara S. Use of thermodynamic data to determine surface tension and viscosity of metallic alloys. MRS Bulletin. 1999;**24**:45-51

[157] Wang L, Chou K-C, Seetharaman S. A comparison of traditional geometrical models and mass triangle model in

calculating the surface tensions of ternary sulphide melts. Calphad. 2008;**32**:49-55

[158] Kaptay G. A method to calculate equilibrium surface phase transition lines in monotectic systems. Calphad. 2005;**29**:56-67

[159] Novakovic R, Giuranno D, Delsante S, Borzone G. Bulk and surface properties of liquid Cr-Nb-Re alloys. Journal of Phase Equilibria and Diffusion. 2014;**35**:445-457

[160] Taskinen J, Yliruusi J. Prediction of physicochemical properties based on neural network modelling. Advanced Drug Delivery Reviews. 2003;**55**: 1163-1183

[161] Qiao Z, Yan L, Cao Z, Xie Y. Surface tension prediction of high-temperature melts. Journal of Alloys and Compounds. 2001;**325**:180-189

[162] Yeum K, Speiser R, Poirier DR. Estimation of the surface tensions of binary liquid alloys. Metallurgical Transactions B. 1989;**20**:693-703

[163] Picha R, Vřešťál J, Kroupa A. Prediction of alloy surface tension using a thermodynamic database. Calphad. 2004;**28**:141-146

[164] Bradshaw R, Warren M, Rogers J, Rathz T, Gangopadhyay A, Kelton K, et al. Containerless measurements of thermophysical properties of $Zr_{54}Ti_8Cu_{20}Al_{10}Ni_8$. Annals of the New York Academy of Sciences. 2006;**1077**: 63-74

[165] Hoffmann M, Wynblatt P. Surface composition of ternary Cu-Ag-Au alloys: Part II. A comparison of experiment with theoretical models. Metallurgical Transactions A. 1991;**22**: 1841-1848

[166] Gallois B, Lupis C. Surface tensions of liquid Ag-Au-Cu alloys. Metallurgical Transactions B. 1981;**12**:679-689

[167] Pajarre R, Koukkari P, Tanaka T, Lee J. Computing surface tensions of binary and ternary alloy systems with the Gibbsian method. Calphad. 2006;**30**: 196-200

[168] Hu J, Chen S, Cai W, Chou K, Zhou G. Predicting the surface tension of ternary by using geometric solution model. Journal of University of Science and Technology Beijing. 1990;**12**: 558-562

[169] Calvo F. Molecular dynamics determination of the surface tension of silver-gold liquid alloys and the Tolman length of nanoalloys. The Journal of Chemical Physics. 2012;**136**:154701

[170] Benisek A, Dachs E. The vibrational and configurational entropy of disordering in Cu_3Au. Journal of Alloys and Compounds. 2015;**632**: 585-590

[171] Muggianu YM, Gambino M, Bros J. Enthalpies of formation of liquid alloys bismuth-gallium-tin at 723k-choice of an analytical representation of integral and partial thermodynamic functions of mixing for this ternary-system. Journal de Chimie Physique et de Physico-Chimie Biologique. 1975;**72**:83-88

[172] Kohler F. Zur Berechnung der thermodynamischen Daten eines ternären systems aus den zugehörigen binären Systemen. Monatshefte für Chemie und verwandte Teile anderer Wissenschaften. 1960;**91**:738-740

[173] Toop G. Predicting ternary activities using binary data. Transactions of the Metallurgical Society of AIME. 1965;**233**:850

[174] Chou K-C. A general solution model for predicting ternary thermodynamic properties. Calphad. 1995;**19**:315-325

[175] Butler J. The thermodynamics of the surfaces of solutions. Proceedings of

the Royal Society of London. Series A, Containing Papers of a Mathematical and Physical Character. 1932;**135**: 348-375

[176] Dogan A, Arslan H, Dogan T. Estimation of excess energies and activity coefficients for the penternary Ni-Cr-Co-Al-Mo system and its subsystems. The Physics of Metals and Metallography. 2015;**116**:544-551

[177] Dogan A, Arslan H. Comparative thermodynamic prediction of integral properties of six component, quaternary, and ternary systems. Metallurgical and Materials Transactions A. 2015;**46**:3753-3760

[178] Arslan H. Determinations of enthalpy and partial molar enthalpy in the alloys Bi-Cd-Ga-In-Zn, Bi-Cd-Ga-Zn and Au-Cu-Sn. Materials Chemistry and Physics. 2015;**153**:384-389

[179] Arslan H, Dogan A. An analytical investigation for thermodynamic properties of the Fe-Cr-Ni-Mg-O system. Russian Journal of Physical Chemistry A. 2015;**89**:180-189

[180] Arslan H, Dogan A, Dogan T. An analytical approach for thermodynamic properties of the six-component systems Ni-Cr-Co-Al-Mo-Ti and their subsystems. The Physics of Metals and Metallography. 2013;**114**:1053-1060

[181] Arslan H. Analytical determination of partial and integral properties of the six components systems Ni-Cr-Co-Al-Mo-Ti and their subsystems. Physica B: Condensed Matter. 2014;**438**:48-52

[182] Dogan A, Arslan H. An investigation on surface tensions of Pb-free solder materials. Philosophical Magazine. 2016;**96**:2887-2901

[183] Arslan H, Dogan A. Thermodynamic investigations on the component dependences of high-entropy alloys. Russian Journal of Physical Chemistry A. 2016;**90**: 2339-2345

[184] Tanaka T, Hack K, Iida T, Hara S. Application of thermodynamic databases to the evaluation of surface tensions of molten alloys, salt mixtures and oxide mixtures. Zeitschrift fuer Metallkunde. 1996;**87**:380-389

[185] Iida T, Guthrie RI. The Physical Properties of Liquid Metals. Oxford, UK: Clarendon Press; 1988

[186] Moser Z, Gasior W, Pstrus J, Zakulski W, Ohnuma I, Liu XJ, et al. Studies of the Ag-In phase diagram and surface tension measurements. Journal of Electronic Materials. 2001;**30**: 1120-1128

[187] Moser Z, Gąsior W, Pstruś J, Dębski A. Wettability studies of Pb-free soldering materials. International Journal of Thermophysics. 2008;**29**: 1974-1986

[188] Kuntz M, Panton B, Wasiur-Rahman S, Zhou Y, Corbin S. An experimental study of transient liquid phase bonding of the ternary Ag-Au-Cu system using differential scanning calorimetry. Metallurgical and Materials Transactions A. 2013;**44**:3708-3720

[189] Prasad L, Mikula A. Role of surface properties on the wettability of Sn-Pb-Bi solder alloys. Journal of Alloys and Compounds. 1999;**282**:279-285

[190] Guggenheim EA. Mixtures: The Theory of the Equilibrium Properties of some Simple Classes of Mixtures Solutions and Alloys. Oxford, UK: Clarendon Press; 1952

[191] Fowler RH, Guggenheim EA. Statistical Thermodynamics: A Version of Statistical Mechanics for Students of Physics and Chemistry. Cambridge: Cambridge University Press; 1960

[192] Singh R. Short-range order and concentration fluctuations in binary

molten alloys. Canadian Journal of Physics. 1987;**65**:309-325

[193] Prasad L, Singh R, Singh V, Singh G. Correlation between bulk and surface properties of AgSn liquid alloys. The Journal of Physical Chemistry B. 1998; **102**:921-926

[194] Bhatia A, Singh R. Short range order and concentration fluctuations in regular and compound forming molten alloys. Physics and Chemistry of Liquids an International Journal. 1982;**11**:285-313

[195] Bhatia A, Singh R. Thermodynamic properties of compound forming molten alloys in a weak interaction approximation. Physics and Chemistry of Liquids an International Journal. 1982;**11**:343-351

[196] Novakovic R, Tanaka T, Muolo M, Lee J, Passerone A. Bulk and surface properties of liquid Ag-X (X = Ti, Hf) compound forming alloys. Surface Science. 2005;**591**:56-69

[197] Kaptay G, Papp Z. On the concentration dependence of the surface tension of liquid metallic alloys theoretical basis. In: Proceedings of Microcad, Materials Science and Technology, University of Miskolc; 2002. pp. 45-50

[198] Hassam S, Ägren J, Gaune-Escard M, Bros J. The Ag-Au-Si system: Experimental and calculated phase diagram. Metallurgical Transactions A. 1990;**21**:1877-1884

[199] Kusoffsky A. Thermodynamic evaluation of the ternary Ag-Au-Cu system—Including a short range order description. Acta Materialia. 2002;**50**: 5139-5145

[200] Sundman B, Fries SG, Oates WA. A thermodynamic assessment of the Au-Cu system. Calphad. 1998;**22**: 335-354

[201] Yan L, Cao Z, Xie Y, Qiao Z. Surface tension calculation of the Ni_3S_2 FeS Cu_2S mattes. Calphad. 2000;**24**: 449-463

[202] Qiao Z-Y, Xing X, Peng M, Mikula A. Thermodynamic criterion for judging the symmetry of ternary systems and criterion applications. Journal of Phase Equilibria. 1996;**17**:502-507

[203] Available from: https://commons.wikimedia.org/wiki/File:Ag-Au-Cu-colours-english.svg