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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

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

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

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:

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

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:

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

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:

$$\sigma^E = \frac{X_1 X_2}{X_{1(12)} X_{2(12)}}\sigma^E{}_{12} + \frac{X_2 X_3}{X_{2(23)} X_{3(23)}}\sigma^E{}_{23} + \frac{X_3 X_1}{X_{3(31)} X_{1(31)}}\sigma^E{}_{31} \tag{11}$$

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:

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

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:

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

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, δ_{ij} = 1 and j ≠ i, δ_{ij} = 0. Tanaka et al. [184] have proposed a model for $G_i^{E,s}$ (T, $X_{j\ (j=2;3,…)}^s$) 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 $p\ln\gamma_i^*$, $q\ln\gamma_i$; 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	$A_{ij}{}^0$	$A_{ij}{}^1$	$A_{ij}{}^2$
Ag-Au	$-128.79 + 0.045836\ T$	$243.86 - 0.060170\ T$	$-455.82 + 0.185348\ T$
	$+0.00680219\ T\ \ln T$	$-0.00965926\ T\ \ln T$	$+0.02786783\ T\ \ln T$
	$-0.00001833\ T^2$	$+0.00002163\ T^2$	$-0.00011934\ T^2$
Ag-Cu	$-982.68 + 0.262677\ T$	$1625.72 - 0.693760\ T$	$-1142.64 - 0.084365\ T$
	$+0.04028137\ T\ \ln T$	$-0.09772514\ T\ \ln T$	$+0.14883052\ T\ \ln T$
	$-0.00013536\ T^2$	$+0.00039080\ T^2$	$-0.00030951\ T^2$
Au-Cu	$94.97 + 0.002865\ T$	$-43.72 - 0.009243\ T$	$-9.72 - 0.008609\ T$
	$-0.00050101\ T\ \ln T$	$-0.00094336\ T\ \ln T$	$-0.00071243\ T\ \ln T$
	$-0.0000217\ T^2$	$+0.00000558\ T^2$	$+0.00002569\ T^2$

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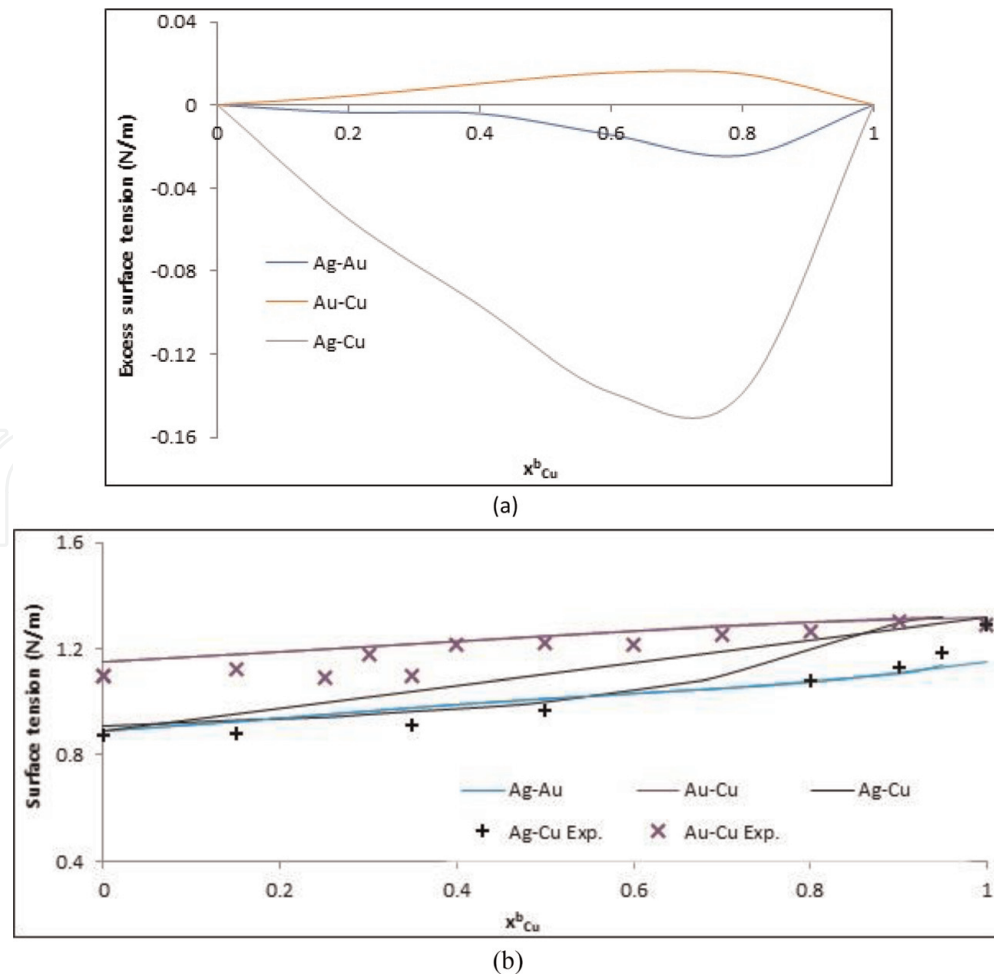
Figure 1.
(a) The excess binary surface tensions σ^E of three sub-binary systems in the Ag-Au-Cu ternary alloy at 1381 K and (b) the binary surface tensions σ of three sub-binary systems in the Ag-Au-Cu ternary alloy at 1381 K.

V_{Au} (m³/mol) = 11.3 (1 + 0.8/10,000 (T/K − 1337.33))/1,000,000	[185]
V_{Ag} (m³/mol) = 11.6 (1 + 0.98/10,000 (T/K − 1234.93))/1,000,000	[46]
V_{Cu} (m³/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, ξ_{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	ξ_{Ag-Au} = 0.934915		ξ_{Au-Cu} = 0.044179		ξ_{Cu-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.
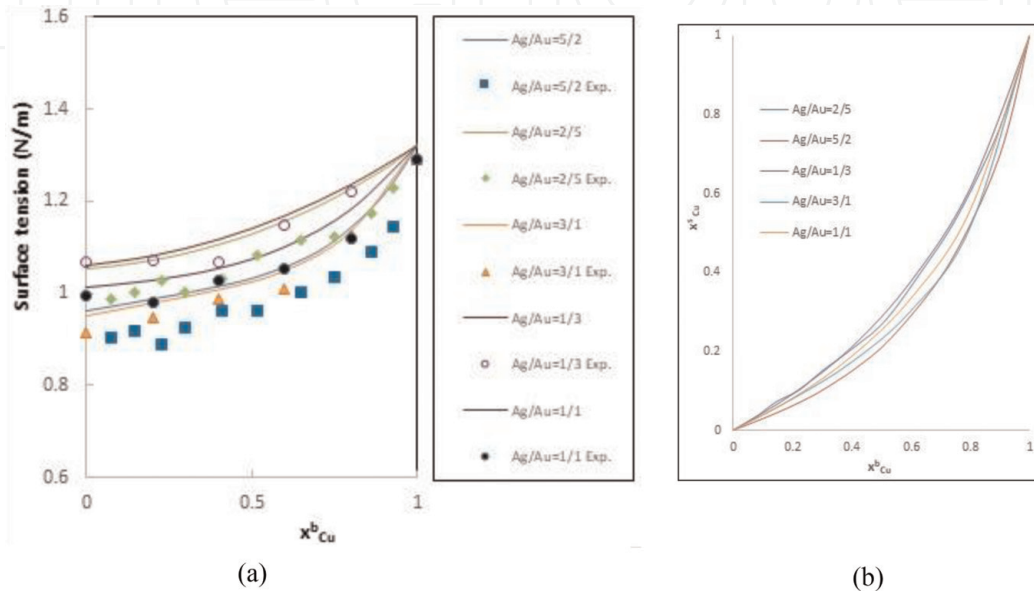
(a)

(b)

(c)

(d)

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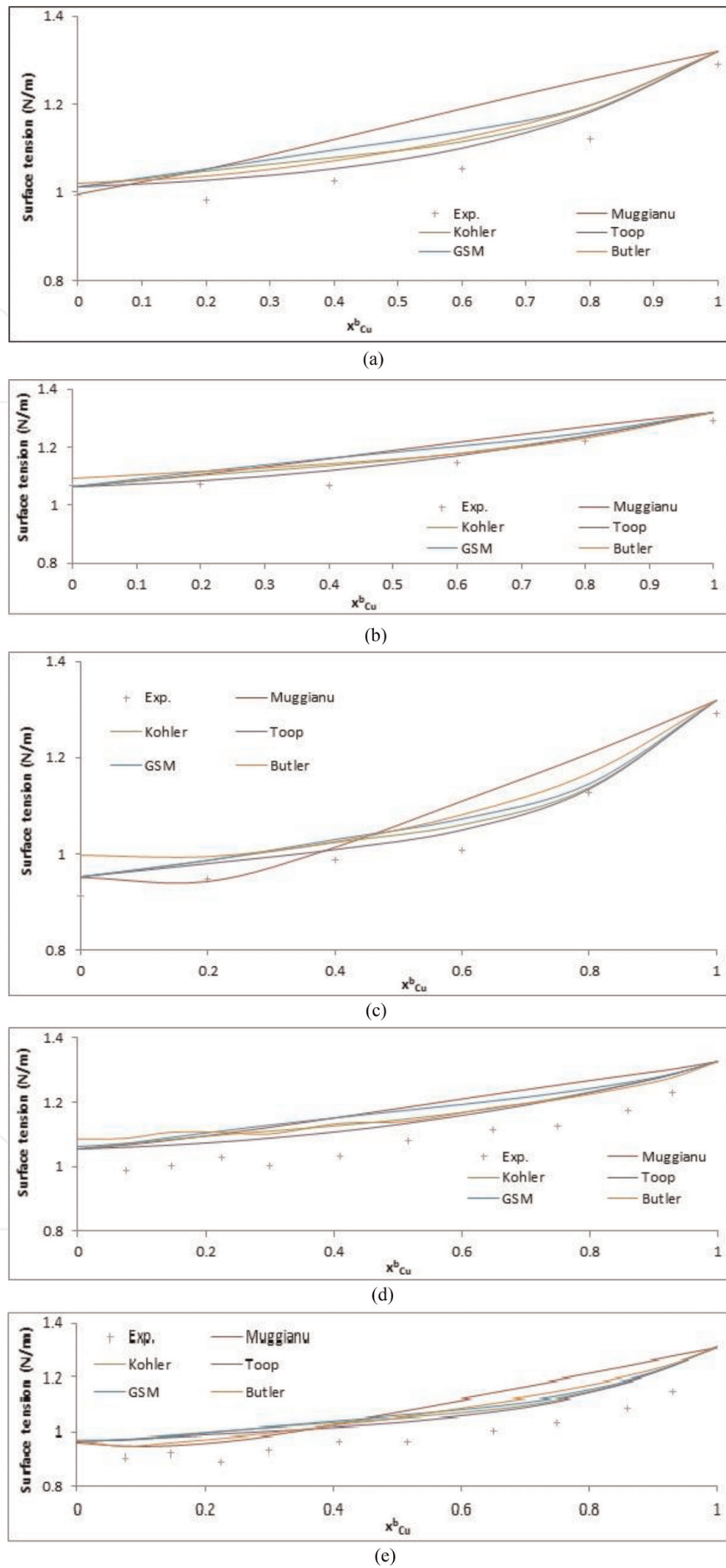
Figure 2.
A comparison of the surface tension values calculated by all the selected models with the experimental values measured in literature [166, 167] for cross section (a) z = x_{Ag}/x_{Au} = 1/3, (b) 1/1, (c) 3/1, (d) 2/5 and (e) 5/2, in the Ag-Au-Cu ternary system.

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 $5/2$).
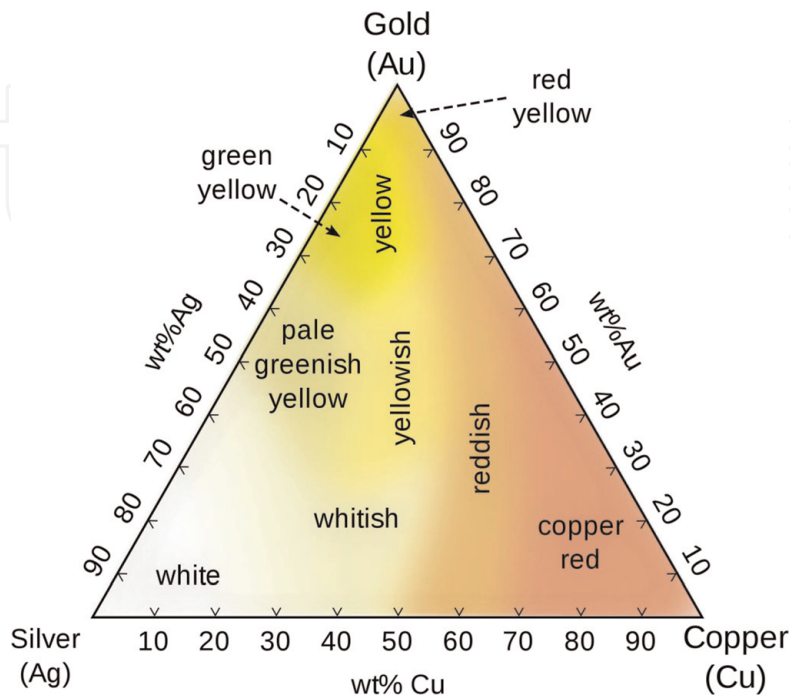
Figure 4.
A phase diagram of the Ag-Au-Cu ternary alloy [203].

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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