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RESEARCH ARTICLE



Investigation of surface tension and viscosity properties of liquid Al–Au binary alloys

Hüseyin Arslan ^{a,b} and Ali Dogan ^a

^aScience and Art Faculty, Department of Physics, Kahramanmaraş Sutcuimam University, Kahramanmaraş, Turkey; ^bInstitute for Graduate Studies in Science and Technology, Department of Material Science and Engineering, Kahramanmaraş Sutcuimam University, Kahramanmaraş, Turkey

ABSTRACT

The experimentally obtained isothermal surface tensions associated with liquid binary alloy Al–Au were obtained as the functions of temperature and composition. The obtained results are compared with theoretical predictions from the Eyring, Guggenheim, Improved Guggenheim, Butler, Ideal Butler, Ideal Solution, Kohler, Egry et al. and Chatain models. The slopes of surface tension with respect to temperature showed negative values. In addition, the experimental data on the viscosity were compared with those from different models, such as Sato, Gasior, Kaptay, Iida-Ueda-Morita, Seetharaman and Sichen, Kozlov-Romanov-Petrov, Kohler, MD (Molecular Dynamic), and Hirai models. The calculated square mean root deviation values of the models are given with respect to both surface tension and viscosity. The best agreement of the surface tensions among chosen models was obtained in the Kohler model for all ranges of Al and Au compositions, respectively.

Les tensions superficielles isothermes expérimentales associées à l'alliage binaire liquide Al–Au ont été obtenues en fonction de la température et de la composition. Les résultats obtenus sont comparés aux prédictions théoriques des modèles de Eyring, Guggenheim, Guggenheim amélioré, Butler, Butler idéal, solution idéale, Kohler, Egry et al. et Chatain. Les pentes de la tension superficielle par rapport à la température ont montré des valeurs négatives. De plus, on a comparé les données expérimentales sur la viscosité à celle de différents modèles, tels les modèles de Sato, Gasior, Kaptay, Iida-Ueda-Morita, Seetharaman et Sichen, Kozlov-Romanov-Petrov, Kohler, MD (dynamique moléculaire) et Hirai. Les valeurs calculées de déviation quadratique moyenne des modèles sont données par rapport à la fois à la tension superficielle et à la viscosité. Le meilleur accord des tensions superficielles parmi les modèles choisis a été obtenu avec le modèle de Kohler pour toutes les gammes de compositions de Al et Au, respectivement.

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1. Introduction

Due to its high tensile strength and low density, Al-based alloy systems are very attractive, especially for aerospace and automotive applications. Moreover, the systems where aluminum is alloyed with transition metals attract attention from material scientists and industrialists due to their high thermal stability. The Al–Au alloy system is an example of eutectic alloy, and its ability to form glass has been recently discussed in the current literature [1]. The thermomechanical properties of Al–Ni systems, such as alloys Al–Ni [2], Al–Fe [2], Al–Ti [3], Al–Cu [4], and Al–Ag [4] are given in the related references, respectively. Al–Cu [4] and Al–Ag [4] as well as Al–Ni [2] and Al–Fe [2] have a strong negative residual volume when the mixture is formed, and their examination in terms of microscopic

structures is given in the reference [5]. Binary Al–Au alloys are also physio-chemically interesting because they combine two elements with opposite properties. When the periodic table is examined, it is seen that Au has a large atomic mass while Al has a low atomic mass. It is also known that the more inert Au is, chemically, highly reactive. Al has a low atomic mass and is chemically highly reactive, while the rather inert Au exhibits a large atomic weight. In addition, the density of liquid Al is much smaller than that of Au, while the molar volume of Al is slightly larger. Therefore, it would be appropriate to apply the perfect solution model for the liquid alloy discussed in this study. Moreover, in order to examine the effect of the intermetallic phases on surface tension, Brillo et al. have compared experimentally determined surface tension data with

those of Butler [6] and Chatains [7,8] for sub-regular solutions. Many references in the reference list in this paper are based on Brillo's surface tension studies, since the articles on surface tension measurements are mostly about the electromagnetic levitation (EML) method. There are two advantages of the EML method. First, the levitate state induced by the rf electromagnetic field can effectively prevent the container wall from being contaminated and increase supercooling. Second, the eddy currents in the sample caused by the inhomogeneous electromagnetic field simultaneously heat the sample and eventually melt it, even for some high melting point alloys. After the 1980s, this method, together with non-contact diagnostic tools, is preferred by most researchers in the study of liquid alloys.

Remarkably, the surface tension and viscosity of Al-Au alloys pair two quantities with approximately the same magnitude at high temperatures [9–19] and viscosity. These thermo physical properties are treated in the present study *via* some models [7,8,20–26], the Butler model [6], and some physical models of viscosity [6,27–35]. On the other hand, recently, viscosity plays an important role in the thermo physical properties of metal melts [36,37] and molecular dynamics simulations of the viscosity [5,38].

It is a well-known fact that many mineral sources have several components. In a metallurgical process, the solutions to be discussed are also known to form a multi-component solution instead of a simple binary solution. It is a demanding and time-consuming task to obtain experimental data concerning viscosity, surface tension, molar volume etc. of the multi-component systems at high temperatures.

To obtain the necessary data, theoretical calculations using various models should be used to predict properties. Recently, due to the simplicity and effectiveness of the geometric models, they have been preferred compared to other physical models. In the meantime, it would be helpful if the readers are given a brief overview of how geometric models work. In geometric models, binary combinations are reached by using geometric relations in an isothermal Gibbs triangle. Typically, the binary compositions are obtained from the intersection of an isogram, passing through the ternary composition of interest with the sides of the triangle. An isogram is a line on a fixed value of a given quantity, such as a mole fraction, a mole fraction ratio, or a mole fraction difference. The binary composition, for example, X_1 , can be expressed in terms of the ternary compositions x_1 , x_2 , and x_3 in the Gibbs triangle. Geometric models used in the excess energy calculation can also be classified as symmetrical and asymmetrical models. These relations between binary and ternary

compositions have different forms for the geometric models in question.

The aim of this study is to make a comparison between the surface tension results obtained from the geometric / physical models and those experimental values. The same procedure was also carried out for the viscosity values. For comparison, a statistical method related to the square mean root deviation was used.

2. Methods

2.1. Some models considered in surface tension and viscosity

Egry model: The expression of the Egry model, which creates a bond between viscosity and surface tension, is given as follows [22]:

$$\lambda/\eta = (15/16)(kT/M)^{1/2} \quad (1)$$

where k , m , and T are Boltzmann constant, atomic mass, and temperature, respectively.

On the other hand, some of the physical models among the viscosity models are given in this chapter briefly, while some of these are referred to in the introduction.

2.2. Some models considered in surface tension

Butler's model: Butler's model is used in order to calculate the surface tension of the mixtures. According to Equation (2), using the density and surface tension of pure metals and thermodynamic data of the liquid binary alloy systems, the surface tension of the system can be written as:

$$\begin{aligned} \gamma &= \gamma_i + \frac{(RT)}{S_i} \ln \frac{(X_i^s)}{(X_i^b)} \\ &+ \frac{1}{S_i} \{G_i^{(E,s)}(T, X_j^s (j=2,3,n)) - G_i^{(E,b)}(T, X_j^b (j=2,3,\dots))\}, i \\ &= 1, 2, 3, \end{aligned} \quad (2)$$

where R , T , γ_i , S_i are gas constant, temperature in K, the surface tension of pure component i , and the surface area of component i , respectively. Moreover, x_i^s and x_i^b are the mole fractions associated with the surface and the bulk phases, respectively. S_i is the molar surface area, which is calculated from the following equation:

$$S_i = 1.091 N_a^{(1/3)} (M_i/\rho_i)^{(2/3)} \quad (3)$$

where N_a , M_i , and ρ_i are denoted as Avogadro's number, atomic mass, and density. $G_i^{E,s}$ and $G_i^{E,b}$ in Equation (1) are the partial excess Gibbs free energies of component i

in the surface and bulk phases, respectively $G_i^{E,b}$. These quantities can be determined or calculated from the data gathered in the existing literature. It is assumed that $G_i^{E,s} = \beta G_i^{E,b}$ for liquid metals [6]. β is a parameter taking into account the reduced coordination at the surface. According to Brillo et al. [39], it is taken as 0.83. Using the following expression, to calculate γ and x_i^s , the expression $\sum_{i=1}^n x_i^s = 1$ can easily be used in the calculation.

Eyring Model: This model is based on the statistical thermodynamic theory of Eyring [40] to calculate the surface tension of binary liquid alloys.

$$\gamma(T) = (kT/\phi)(V_s/V)^2 \left[\frac{E_s(1-3f)}{RT} + 0.75 \ln 0.75(1+f) \right] \quad (4)$$

where the atomic area is defined as $\phi = \left(\frac{\sqrt{3}}{2} \right) \left(\sqrt{2} \frac{V_s}{N_a} \right)^{2/3}$. ϕ , V , and V_s in values used in the calculation of the surface tension of a liquid binary alloy can be written as, respectively: $V(\text{alloy}) = V_1\phi_1 + V_2\phi_2$, $V_s(\text{alloy}) = X_1V_{s1} + X_2V_{s2}$ and $\phi(\text{alloy}) = X_1\phi_1 + X_2\phi_2$

The dissociation energy of an alloy is given by [41]:

$$E_s = x_1^2 E_{s1} + x_2^2 E_{s2} + 2x_1x_2(E_{s1}E_{s2})^{(1/2)} \quad (5)$$

The parameters in this model can be denoted as follows: $V_s = M/\rho(T)$ and $V = M/\rho(T)$ are the molar volumes at the melting point T_L (K) and any temperature T (K) of liquid metal alloy, N_a is Avogadro's number, E_s is the sublimation energy, f is a constant determined numerically *via* the fitting procedure, k is the Boltzmann factor, and M is the atomic weight.

Guggenheim model: Guggenheim proposed an expression concerning surface tension of the binary alloys. This expression can be written as [23]:

$$\exp\left(-\frac{\gamma A}{kT}\right) = X_1 \exp\left(-\frac{\gamma_1 A_1}{kT}\right) + \exp\left(-\frac{\gamma_2 A_2}{kT}\right) \quad (6)$$

where γ is the surface tension of the binary alloy, γ_1 and γ_2 are surface tensions of the individual components of the alloy, and A is the molar surface area of the binary alloy defined by:

$$A = f^l N_a^{1/3} V_i^{2/3} \quad (7)$$

f^l is the atomic arrangement factor for the liquid surface and $A_1 = A_2 = A_3 = A(\text{alloy})$. The density of the liquid alloys can be written using the molar volume of pure Al and Au, which is defined as:

$$V_i = M_i/\rho_i \quad (8)$$

and

$$\rho_{\text{alloy}} = \frac{X_1 M_1 + X_2 M_2}{X_1 V_1 + X_2 V_2 + V^E} \quad (9)$$

where ρ and ρ_i are the densities of liquid alloy and pure component i , respectively. M and M_i are atomic weights of the liquid alloy and pure component i , respectively.

Improved Guggenheim model: A modification of the Guggenheim theory is obtained by replacing volume fractions v_1 and v_2 in the place of the mole fractions as follows [24]:

$$\exp\left(-\frac{\gamma A}{kT}\right) = v_1 \exp\left(-\frac{\gamma_1 A_1}{kT}\right) + v_2 \exp\left(-\frac{\gamma_2 A_2}{kT}\right) \quad (10)$$

where

$$v_1 = \frac{V_1 X_1}{V_1 X_1 + V_2 X_2}, v_2 = 1 - v_1, A = V_1 v_1 + V_2 v_2, A_i = V_i^{2/3} = (M_i/\rho_i)^{2/3} \quad (11)$$

Chatain model: Because it cannot make statements about the concentration gradient vertical to the surface in liquid alloys, Chatain or multilayer model – considering the surface as a stack of k parallel layers where the atoms are thought to reside on regular lattice sites – was developed. The surface tension γ of an A–B binary liquid may be written as the sum of three contributions [42]:

$$\gamma = U^s \psi^{-1} - T S^s \psi^{-1} - (\mu_A \Gamma_A + \mu_B \Gamma_B) \quad (12)$$

where U^s and S^s denote the internal energy of the surface and its entropy, respectively. μ_i is the chemical potential of component i in the solution and Γ_i denotes the so-called adsorption, i.e. the surface excess number of moles of the component i , per unit area Ψ . The Chatain model can be applied to systems obeying the subregular solution model with a maximum of two Redlich–Kister parameters, including A_{ij}^0 and A_{ij}^1 . To calculate the internal energy term, the energies of the central atoms of kind A or B, i.e. $E_A(p)$ or $E_B(p)$, respectively, can be taken as parabolic functions of the numbers of the respective foreign neighbours, p or q :

$$E_A(p) - E_A^0 = \frac{A_{ij}^0 (z-1) - A_{ij}^1 (3z-1)}{z(z-1)(3z-1)} p(2z-p) \quad (13)$$

$$E_B(p) - E_B^0 = \frac{A_{ij}^0 (z-1) + A_{ij}^1 (3z-1)}{z(z-1)(3z-1)} q(2z-q) \quad (14)$$

γ is expressed as the sum of six terms with the entropy of the mixture [8] assumed as ideal:

$$\gamma \psi = \text{minimized}(\gamma_1 + \gamma_2 + \gamma_3 + \gamma_4 + \gamma_5 + \gamma_6) \psi \quad (15)$$

γ_1 includes all contributions to the internal energy from the first, i.e. topmost monolayer ($n = 1$), and γ_2 is the contributions from the last layer for which $n = k$. The expressions of other terms are found in Ref. [8]. In the last equation, the surface area ψ is approximated by the average of Ψ_{Al} and Ψ_{Au} . The atoms lie on lattice sites, and their total coordination number, Z , is split into a lateral coordination number; z_l , is neighbours lying in the atomic layers parallel to the surface, and an interlayer coordination number; z_v , is perpendicular to the surface. These coordination numbers are related as follows:

$$z = z_l + 2 z_v \quad (16)$$

The six terms γ_i depend on the surface tensions of the pure components, the compositions $x_i^{(n)}$ of a layer n (i.e. $1 < n < k$), and its neighbouring ones as well as the coordination numbers, z , z_l and z_v . $z_v = 6$ is the lateral coordination number in the layers parallel to the surface. $z_v = 3$ is the number of neighbours of an atom in an adjacent atomic layer and the total coordination number, $z = z_l + 2 z_v$ is Equation (16). The equation concerning $\gamma\psi$ mentioned above has to be calculated layer-by-layer and the global minimisation is performed by a Monte Carlo algorithm.

2.3. Some models considered in viscosity

Muggianu Model: The expression for viscosity deviation of this model [20] concerning the binary mixture is given in the closed form as:

$$\eta = \frac{4x_1x_2}{(1+x_1-x_2)(1+x_2-x_1)} \eta_{12} \left(\frac{(1+x_1-x_2)}{2} ; \frac{(1+x_2-x_1)}{2} \right) \quad (17)$$

Kohler model: This model, which has a symmetrical character for the binary mixture [21] is as follows:

$$\eta = (x_1 + x_2)^2 \eta_{12} \left(\frac{x_1}{x_1 + x_2} ; \frac{x_2}{x_1 + x_2} \right) \quad (18)$$

When the excess thermodynamic properties of the three binary subsystems are similar to each other, it is said that the ternary system is symmetric.

Budai-Benko-Kaptay Model: The viscosity of a multi-component liquid alloy has been derived by Budai-Benko-Kaptay [27] as follows:

$$\eta = A \frac{(\sum_i x_i M_i)^{1/2}}{(\sum_i x_i V_i + \Delta V^E)^{2/3}} T^{1/2} \exp \left[\frac{B}{T} \left(\sum_i x_i T_{m,i}^* - \frac{\Delta H}{Q.R} \right) \right] \quad (19)$$

Here, the expression of the effective melting points is give as follows:

$$T_{m,i}^* = \frac{T}{B} \ln \left[\frac{\eta_i V_i^{(2/3)}}{A M_i^{(1/2)} T^{(1/2)}} \right] \quad (20)$$

where, M_i and V_i are atomic mass and molar volume of the corresponding metal i , respectively. The integral enthalpy of mixing is ΔH . X_i is the concentration of component i and the value of Q is 25.4. For the sake of simplicity, the term ΔV^E is the excess molar volume of the mixing, which has a negligible quantity. $T_{m,i}^*$ is defined as an effective melting point of the pure component. The values of A and B in the equation given above are taken as $(1.80 \pm 0.39) \cdot 10^{-8} \text{ (J/Kmol}^{1/3})^{1/2}$ and 2.34 ± 0.20 , respectively.

Kozlov-Romanov-Petrov: By considering the atomic vibration frequencies to describe the viscosity of a liquid, Kozlov-Romanov-Petrov equation can be expressed as [28]:

$$\ln \eta = \sum_{i=1}^N x_i \ln \eta_i - \frac{\Delta H_{mix}}{3RT} \quad (21)$$

where ΔH_{mix} represents the integral enthalpy of mixing, η_i and x_i are the viscosity and concentration of the given component i , respectively, T and R are the absolute temperature and universal gas constant, respectively.

Sato Model: In this model, the logarithmic additivity was considered to be a good criterion for representing the composition dependence of viscosity [30]:

$$\log \eta_{alloy} = \sum_{i=1}^N x_i \log \eta_i^0 + \sum_{i=1}^N x_i \frac{E_i}{2.3RT} \quad (22)$$

η_i^0 and E_i in Equation (11), are the pre-exponential factor and activation energy of the viscous flow.

Moelwyn-Hughes Model: Moelwyn-Hughes developed a simple model [34] to set up the composition dependence of viscosity for a binary system, so that its expression is given by:

$$\eta = \sum_{i=1}^N \eta_i c_i - (1 - 2 \frac{\Delta H_{mix}}{RT}) \quad (23)$$

Where η_i and c_i are viscosities of elements, and the mole fractions, respectively, and Ω is the regular solution interaction parameter. The model seems to predict the viscosity of some metal systems successfully, whereas it fails to give a prediction of the low viscosity behaviour at the eutectic compositions. The corrections for the atomic size are proven to be necessary.

Schick et al.: Recently, a new model has been proposed to determine the viscosity of Al-Cu liquid alloys

by Schick *et al.* [29]. An equation of it can be given as follows:

$$\ln \eta = \sum_{i=1}^N x_i \ln x_i \eta_{i,\infty} + \frac{\sum_{i=1}^N x_i E_i - \Delta H_{mix}}{RT} \quad (24)$$

where E_i and $\eta_{i,\infty}$ represents the activation energy and pre-exponential factor of viscous flow, respectively.

Hirai Model: Up until now, the use of the Hirai model [36] to predict liquid metal viscosity has been relatively satisfactory. It can be written as follows:

$$\eta = 1.7 \cdot 10^{-7} \frac{\rho^{2/3} T_L^{1/2}}{M^{1/6}} \exp\left[\frac{2.65 T_L^{1.27}}{R} \left(\frac{1}{T} - \frac{1}{T_L}\right)\right] \quad (25)$$

Seetharaman–Du Sichen Model: The Seetharaman–Du Sichen model [33] links the activation energy to the excess Gibbs energy G^E . It is known that this model is only valid for binary systems. The following semiempirical expression can also be given for a multi-component system:

$$\eta = \frac{h N_a}{V} \exp \frac{\sum_i^n c_i \Delta G_i^* + 3RT \sum_{i=1}^n \sum_{j>i}^{n+1} c_i c_j + RT \sum_{i=1}^n c_i \ln c_i + G^E(c_1, c_2, \dots, c_n)}{RT} \quad (26)$$

where ΔG_i^* is calculated from the equation given by:

$$\Delta G_i^* = RT \ln \left(\frac{\eta_i V_i}{h N_a} \right) \quad (27)$$

where h is Planck's constant. N_a is Avogadro's number, V_i is molar volumes of components, and ΔG^* represents the Gibbs activation energy for viscosity.

Kaptay Model: The Kaptay equation [32] relates the activation energy to the enthalpy of mixing ΔH_{mix} . It is carried out by way of introducing a semi empirical parameter, $\alpha = 0.155$. This parameter can be estimated from the properties of pure components in the alloys:

$$\eta = \frac{h N_a}{V} \exp \frac{\sum_i^n c_i \Delta d^2 G_i^* - \alpha \Delta H_{mix}}{RT} \quad (28)$$

where N_a , c_i , V , and h denote Avogadro's number, the mole fractions, molar volume, and Planck's constant, respectively.

Gasior Model: The biggest difference from other models is that in the Gasior model, the viscosity of the alloy is based on the excess entropy in the system. The new simpler model was proposed by this researcher [35] as follows:

$$\eta = \sum_{i=1}^N \eta_i c_i - \left(1 - 2 \frac{S^E}{R}\right) \quad (29)$$

where η_i , c_i , and R are viscosities of the elements, mole fractions, and the universal gas constant, respectively.

Ueda and Morita: Ueda and Morita have presented the following model [31] given by the equation as follows:

$$\eta = (X_1 \eta_1 + X_2 \eta_2) \left\{ 2 \left[1 + \frac{X_1 X_2 (M_1^{1/2} - M_2^{1/2})^2}{(X_1 M_1^{1/2} + X_2 M_2^{1/2})^2} \right]^{(1/2)} - 1 - \frac{5 X_1 X_2 (d_1 - d_2)^2}{(X_1 d_1^2 + X_2 d_2^2)} - \Delta \right\} \quad (30)$$

where

$$\Delta = 0.12 \frac{\Delta H_{mix}}{RT} \text{ or } \Delta = 0.12 \frac{\Delta G^E}{RT}$$

3. Results and discussion

Describing the temperature dependence of surface tension of the pure elements, both theoretical and exper-

imental studies suggest that there is a well-known universal linear relationship in the following form:

$$\gamma(T) = \gamma_L + \gamma_T (T - T_L) \quad (31)$$

Moreover, similar to this expression, it is also possible to write an equation for the density of the alloy as given:

$$\rho(T) = \rho_L + \rho_T (T - T_L) \quad (32)$$

In Equation (33) and (34), γ_L and ρ_L are the surface tension and density at the liquid's temperature, γ_T and ρ_T are the temperature coefficients associated with the surface tension and density. The linear relationship we just mentioned for the surface tension is valid for some binary alloys. When most liquids are examined in the laboratory, it can be understood that surface tension shows negative temperature dependence. A few exceptions can be given, which exhibit a positive temperature coefficient. The temperature dependence of the surface tension has been reported for a few Cu-Si systems by some investigators, when the composition exceeds 30% from negative to positive values [43]. Similar conditions have also been reported in Zr-based binary and ternary alloys. This can be explained by the effect of local ordering structures such as clusters of intermetallic compounds in the phase diagram on thermodynamic properties. It should be noted that these effects on the surface

Table 1. Parameters of liquid Al and Au contents [4, 47].

System	T_L (K)	ρ_T (10^{-4} g / cm ³ K)	ρ_L (g / cm ³)	Es (kJ/mol)	Vs (cm ³ /mol)	ϕ (cm ² /atom) $\times 10^{-16}$	Refs.
Al	933.5	−2	2.35	332.351	11.5	7777	[4]
Au	1336	−11	17.4	368.000	11.32	7.7	[47]

properties in liquids cannot be neglected. As observed in pure liquid metals, the calculated surface tension of the binary alloys showed a great deviation from the experimental results. For instance, the surface tension of the Ni₅₀Al₅₀ alloy in the range of 1900–2000 K temperature range was estimated to be as low as nearly 33–35% from the current experimental values [44]. On the other hand, it is reported that the calculated values of the binary alloys Ni-Cu were nearly 30–40% higher than the experimental data [45].

In this study, the physical properties of these alloys were investigated by applying a large number of physical models to the binary liquid alloy systems for surface tension and viscosity at 1400 K. Al-Au alloy and its components are in the liquid state at the given temperature. This temperature is above the melting temperatures of the components. Therefore, the alloy temperature was selected as 1400 K in this study. The surface tension values of alloy components in a summary of the introduction are used as input data for calculations in a large number of surface tension models. However, there are also several experimental studies on changes in the value of these quantities due to reasons such as oxidation and contamination [46], when measured at high temperatures. Therefore, there is a model which has a statistical structure, which is one of the surface tension models examined recently. Only the Eyring model, which is a physical model that requires the densities of alloy components, melting temperature values, molar volume, and sublimation energy values, is used. These data are physical ones and can be easily found in the literature. The experimental literature data on the density and its temperature dependence of pure liquid Al, Au in Al-Au alloys in the present work were employed for the volumes at any given temperature in Table 1 [4, 47].

On the other hand, the required Redlich–Kister parameters concerning the excess Gibbs energy for the Butler calculations associated with the liquid binary alloys Al–Au [48] are given in Table 3.

The Kohler model, which is one of the symmetrical geometric models, has a simple and useful mathematical expression. Therefore, it is included in this study because it gives very suitable results for surface tension and viscosity values. So, the measured surface tension values of Al-Au binary alloys in the calculation of the

excess surface tension of the binary alloy system were fitted to the Redlich–Kister equation [49].

$$Z^{Exc} = x(1-x) \sum_{k=0}^n A_{ij}^k (1-x)^k \quad (33)$$

where Z^{Exc} denotes γ^{Exc} or η^{Exc} and n is the optimal number of parameters (in this work, n is taken as 5). Table 4 displays the calculated Redlich–Kister parameters of the excess surface tension (mN/m) at 1400 K for the liquid alloys Al-Au. Using the parameters in Tables 1–4, the isothermal surface tension for all models was determined at a constant temperature of 1400 K. The isothermal surface tension values for the calculated and reported experimental data of the surface tension of the liquid alloys Al-Au at 1400 K are tabulated in Table 5.

To compare models, one can calculate the root mean square deviation, S , corresponding to the experimental results for each model. Its equation can be written as:

$$S = \frac{1}{n} \sqrt{\sum_{i=1}^n (\lambda_{calc,i} - \lambda_{exp,i})^2} \quad (34)$$

where $\lambda_{calc,i}$ and $\lambda_{exp,i}$ represent the calculated and experimental viscosities of alloys. Here, n is the total number of experiments concerning the alloys treated in this study. For this purpose, the calculated surface

Table 2. The parameters γ_L and γ_T for pure Al and Au and related to the results taken from the literature [39].

	γ_L (N/m)	γ_T 10^{-4} (N/mK)	refs.
Al	0.866 ± 0.03	-1.46 ± 0.4	[57]
Au	1.140 ± 0.06	-1.83 ± 0.4	[57]

Table 3. Redlich–Kister parameters required for the excess Gibbs free energy associated with the liquid binary alloys Al–Au [48].

Redlich–Kister Parameters (in terms of J / mol)		
${}^0L_{Al-Au}$	−131,996.19	36.42 T (in terms of K)
${}^1L_{Al-Au}$	40,781	−1.896 T (in terms of K)

Table 4. The calculated Redlich–Kister parameters of the excess surface tension (mN/m) at 1400 K concerning the liquid alloys Al-Au.

A_{ij}^0	A_{ij}^1	A_{ij}^2	A_{ij}^3
12	−417.25	178.5	291.75

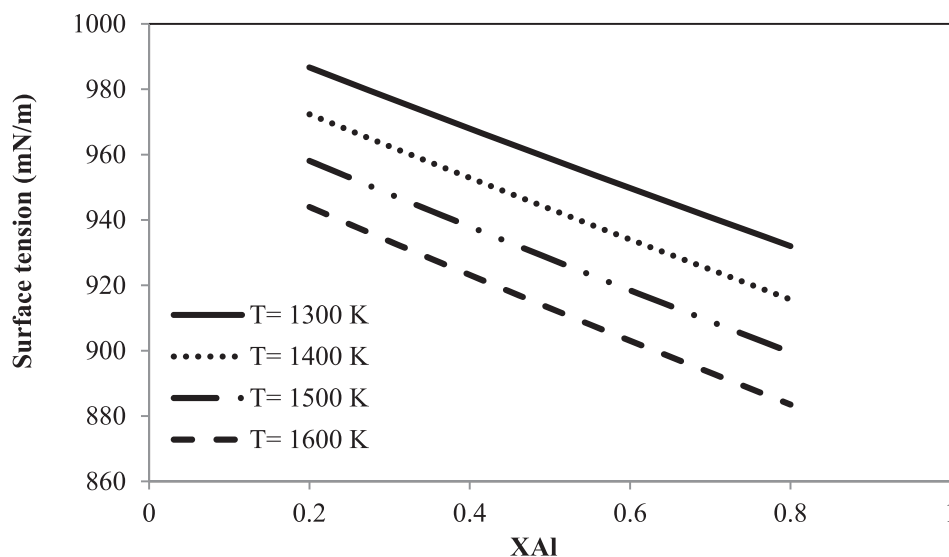
Table 5. Composition dependence of Al for the calculated and reported experimental of the surface tension of the liquid alloys Al–Au at 1400 K.

Al	Eyring	Guggenheim	Butler	Improved Guggenheim	Ideal Butler	Ideal Solution	Kohler	Egry et al	Chatain	Exp.
0	1081.6	1133.3	1112.6	1102.7	1128.5	1128.5	1128.5	1338.8	1112.6	1120
0.15	1051.6	1044.3	1118.5	1023.2	1077.3	992.8	1115.8	-	1081.5	1111.1
0.20	1040.3	1017.2	1114.1	998.6	1057.8	960.4	1101.9	1171.6	1065	1108.9
0.28	1026.9	988.2	1094.1	972.2	1034.4	929.5	1077.7	-	1034.1	1055.6
0.33	1017.2	969	1085.9	954.75	1017.3	910.9	1055.3	955.6	1014.8	1040.7
0.47	991.96	924.6	1037	914.1	972.5	873.1	985.2	-	965.9	1022.2
0.51	983.4	911.1	1014.8	901.7	957.1	870.8	959.9	1013.7	943.7	948.2
0.55	976.4	900.4	985.19	891.9	944.5	868.8	939.5	-	932	918.5
0.69	951.9	865.9	911.1	860.4	899.8	845.5	879.4	711.6	887.4	881.5
0.80	931.6	840	844.4	836.6	861.9	827.5	845.8	682.9	852	844.4
0.87	919.6	825.7	835.6	823.5	839.2	817.4	831.4	-	829.6	835.6
1	897.9	801.4	800	801.4	798	797.9	798	545.6	800	800
S-values	17.8	15.4	9.7	18.6	7.8	79.7	4.4	35.7	69.5	

Notes: The calculated square mean root deviation values of the models are given as S-values at the bottom of the table. The experimental data are taken from Ref. [39].

tension values and root mean square deviations values were calculated to determine which of these models corresponds better with the surface tension results, and S-values associated with those models are also given at the bottom of Table 5. It was concluded that the proposed Kohler model is also suitable for explaining the viscosity results of the Al–Au alloys – although some advantages of the Eyring model among the other models are mentioned above, and the Butler model is the most successful model for testing surface tension values in multicomponent alloys, in which, the surface tension data presented in the work of literature [39] on pure Al deviate at liquid temperature by roughly 15% from those measured under oxygen-reduced conditions [50–53]. Generally speaking, as obviously seen from Table 5, all models are in quantitative agreement with the observed isothermal surface tension. The calculated

compositional dependence of the surface tension of liquid Al–Au alloys for different temperatures as a function of the mole fractions of Al is given in Figure 1, while the surface tension of the liquid alloys for different Al mole fractions as a function of the temperature is given in Figure 2. The calculation results given in the graph are carried out by the Eyring model in the present study. Moreover, Figure 3 shows the calculated surface tension of liquid Al–Au alloys having different Al mole fractions for different temperatures along with the experimental results carried out for different temperatures. It is seen from Figure 3 that the calculations for $0.3 < X_{\text{Al}} < 0.7$ rather agree with the experimental results. Moreover, Figure 4 shows a comparison of the surface tension of liquid Al–Au alloy as a function of the Au content for the two models, such as Eyring and Egry model calculated from the viscosity

**Figure 1.** Compositional dependence of the surface tension of liquid Al–Au alloys for different temperatures as a function of the mole fractions of Al. The calculation results given in the graph are carried out by the Eyring model.

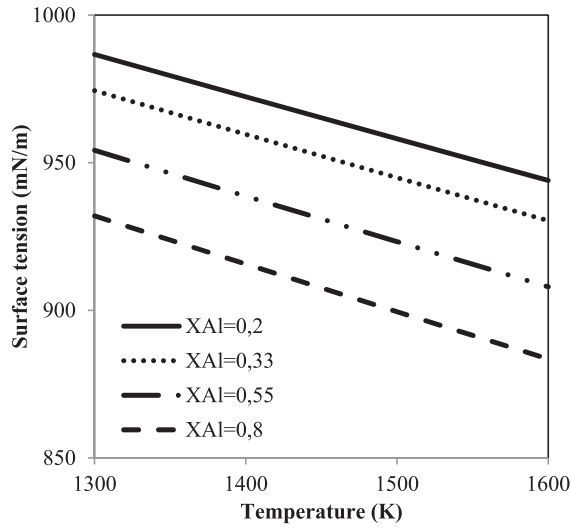


Figure 2. Surface tension of liquid Al–Au alloys for different Al mole fractions as a function of the temperature. The calculation results given in the graph are carried out by the Eyring model.

experiments along with the experimental results at a constant temperature 1400 K. On the whole of the diagram, both calculations agree with each other; the results obtained from Eyring model are in quantitative agreement with the experimental results from the entire range of the Au content. The surface tension curves of liquid Al–Au alloys as a function of the Al bulk concentration at 1400 K, in which, the Kohler model is

considered along with the experimental results are given in Figure 5. While, in Figure 5, no sign of compound formation is seen in the surface tension values around $X_{Al} = 67$ at.% which corresponds to the Al_2Au phase (The $AuAl_2$ intermetallic is referred to as purple gold and feared for its brittleness and low conductivity), a weakly pronounced shoulder is exhibited at $x_{Al} = 42.5$ at % which corresponds to the composition of the less dominant $AlAu_2$ phase [39]. It is reported that such a shoulder at $x_{Al} = 42.5$ at is also exhibited in the surface tension of the chemically similar system Al–Cu [54–56]. In this work, the Kohler model, which may show rather good trend with the shoulders associated with the intermetallic phases just mentioned above because it fits best to the experimental data [39], is shown in Figure 5 together with the experimental results. Figure 6 shows the temperature coefficient of liquid Al–Au alloys as a function of the Al composition x_{Al} along with the results obtained from some models and the experimental results at 1400 K; it is seen from Figure 6 that a slight mutual agreement is observed among the results of the models related to the temperature coefficients of the liquid Al–Au alloys, while Chatain model display similar trends with those of the experimental results [39].

As to viscosity; carrying out similar calculations to the surface tension models considered, the obtained isothermal viscosity values at 1400 K are shown in Table 9

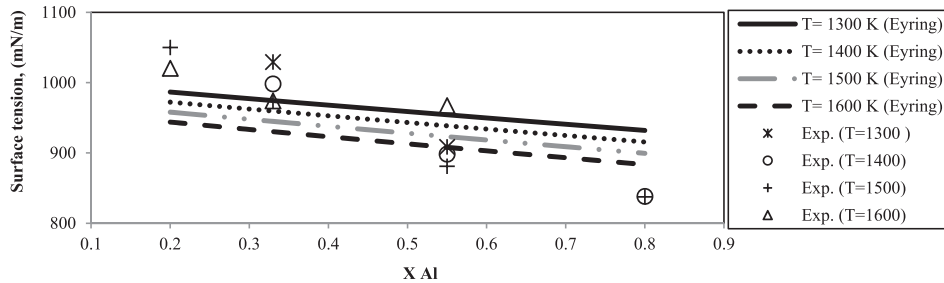


Figure 3. Surface tension of liquid Al–Au alloys with different Al mole fractions for different temperatures along with the experimental results performed for different temperatures. The experimental data are taken from Ref. [39].



Figure 4. Surface tension of liquid Al–Au alloy as a function of the Au content for two models along with the experimental results at 1400 K. The experimental data are taken from Ref. [39].

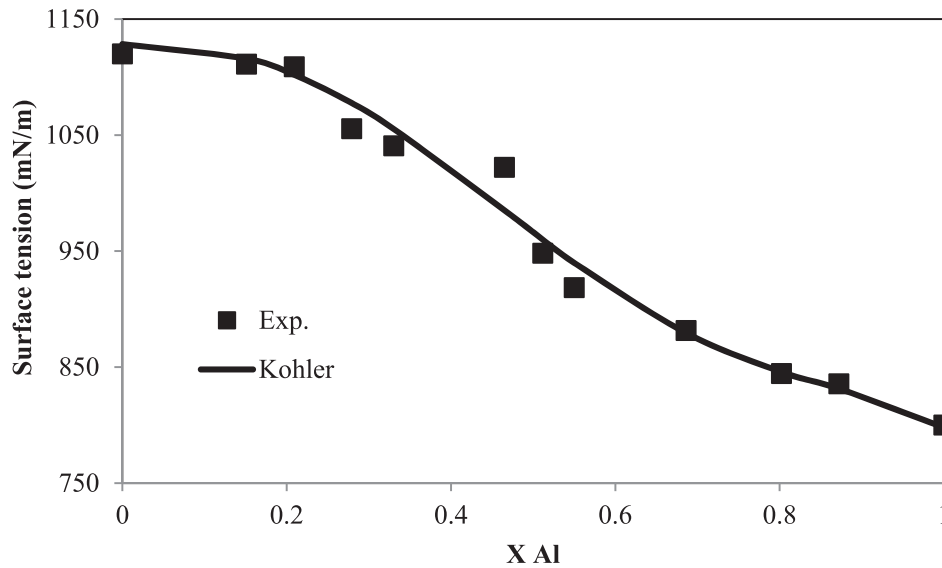


Figure 5. Surface tension of liquid Al–Au alloys as a function of the Al bulk concentration at 1400 K, in which, the Kohler model is considered along with the experimental results. The experimental data are taken from Ref. [39].

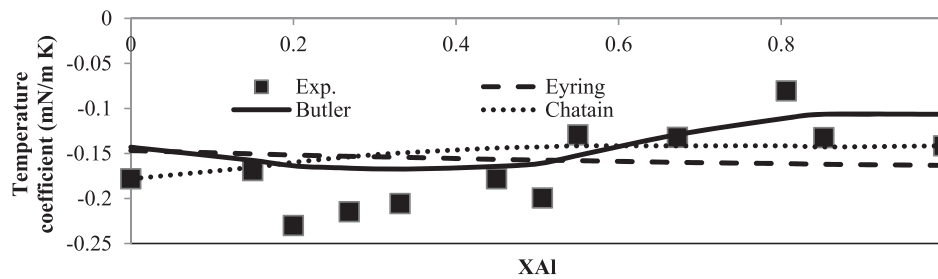


Figure 6. Temperature coefficient of liquid Al–Au alloys as a function of the Al composition x_{Al} at 1400 K along with some models and the experimental results at 1400 K. The experimental data are taken from Ref. [39].

as functions of the Au mole fraction using the related parameters in Tables 6–8. The calculated square mean root deviation values of the models are given as S-values

Table 6. Redlich–Kister parameters (J/mol) required for the enthalpy in the Liquid alloys Al–Au [57].

A_{ij}^0	$-110319.42 - 141.11355 T + 12.825 (T \ln T)$
A_{ij}^1	$32,465.84 + 2.45897 T$
A_{ij}^2	23,553.16

Table 7. The calculated Redlich–Kister parameters of the excess viscosity (mPa s) at 1400 K concerning the liquid alloys Al–Au.

A_{ij}^0	A_{ij}^1	A_{ij}^2	A_{ij}^3
0.088	−0.92	−11.55	8.89

Table 8. Parameters η_∞ and E_A obtained experimentally [5].

Component	η_∞ (mPa s)	E_A (kJ/mol)
Al	0.214	14.3
Au	0.811	23.1

at the bottom of the same table. It is concluded from the results obtained from the calculated viscosities that the proposed Kohler model is also suitable to explain the viscosity results of Al–Au alloy. Therefore, the isothermal viscosity values are redrawn in Figure 7 as functions of the Au mole fraction using the Kohler model. Generally speaking, it can be seen that the viscosity increases monotonically with the increase in Au composition. In addition, the experimental viscosity [5] results and the values obtained from the Kohler model are in good agreement (Table 9). The surface tension and viscosity graphs drawn at 1400 K are also given in Figures 8 and 9, to compare the models with the experimental results.

In this study, the surface tension values of the binary liquid Al–Au alloy were calculated using a large number of physical models at various temperatures – as a function of both Al and Au compositions in a wide temperature range – and compared with the experimental results. In addition, the viscosity values were calculated

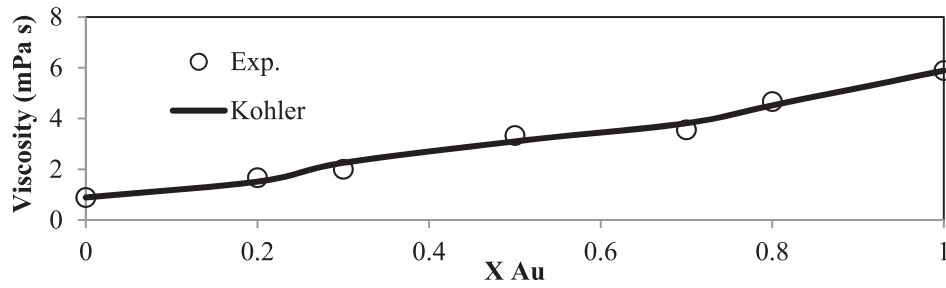


Figure 7. Isothermal viscosity values of Al-Au melts obtained from the experiment and Kohler model at 1400 K. The experimental data are taken from Ref. [5].

Table 9. Composition dependence of Au for the calculated and reported experimental of the viscosity associated with the liquid alloys Al-Au at 1400 K. The calculated square mean root deviation values of the models are given as S-values at the bottom of the table [5].

X_{Au}	Sato et al.	MD	Gasior	Kaptay	Iida-Ueda-Morita	Hirai	Seetharaman and Sichen	Kozlov-Romanov-Petrov	Kohler	Exp.
0	0,708	0,889	0,888	1,146	0,888	1,220	0,901	0,862	0,888	0,888
0,2	1,078	1,444	4,112	1,772	1,901	2,067	0,600	2,006	1,511	1,666
0,3	1,329	-	8,690	2,264	2,337	2,452	0,512	3,526	2,253	2
0,5	2,022	-	17,48	3,570	3,256	3,211	0,443	8,004	3,096	3,333
0,7	3,076	-	15,68	5,161	4,325	3,988	0,619	8,966	3,814	3,555
0,8	3,794	4,467	10,28	5,917	4,887	4,391	0,974	7,645	4,517	4,666
1	5,772	5,222	5,889	6,854	5,888	5,233	6,021	5,760	5,888	5,888
S-values	0,291	1,329	2,961	0,328	0,129	0,155	0,724	1,128	0,069	

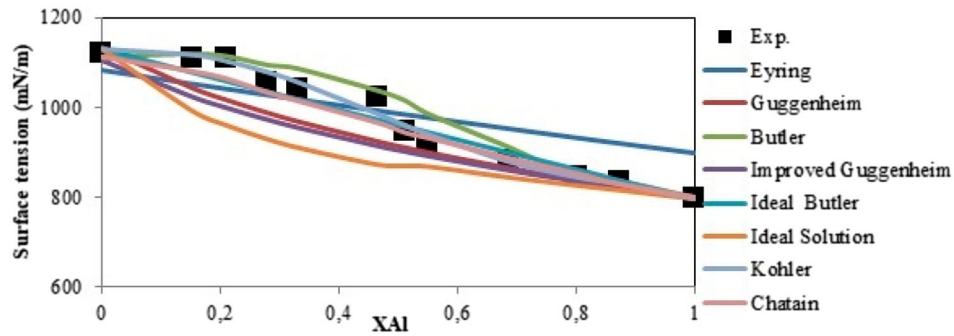


Figure 8. Surface tension of liquid Al-Au alloys as a function of the Al bulk concentration x_{Al} at 1400 K along with the experimental results at this temperature. The experimental data are taken from Ref. [39].

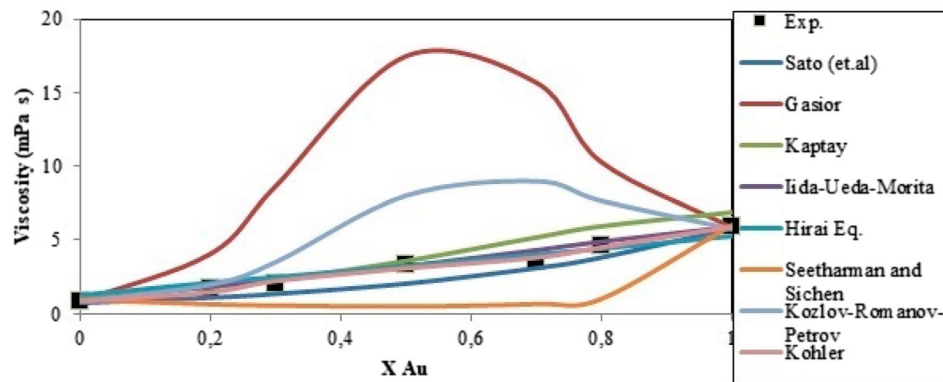


Figure 9. Viscosity of liquid Al-Au alloys as a function of the Au bulk concentration x_{Au} at 1400 K along with the experimental results at this temperature. The experimental data are taken from Ref. [5].

at 1400 K using a large number of physical models as a function of the Au composition and compared with the experimental results covering the entire concentration. It can be seen from the root mean square deviation values calculated for the models in the present study that the Kohler model gives the minimum values. Here is the result that may be important in this study: Except for both the ideal solution and Chatain, the other models are able to reproduce the experimental data sufficiently, but the best agreement of the surface tension and viscosity among chosen models was obtained in the Kohler model – for both surface tension and viscosity in the entire ranges of Al and Au compositions, respectively. According to the authors, this model is good enough to be predictable for presenting the data concerning both the surface tension and viscosity of the liquid binary Al–Au system at 1400 K.

Disclosure statement

No potential conflict of interest was reported by the author(s).

ORCID

Hüseyin Arslan  <http://orcid.org/0000-0003-1509-7725>

Ali Dogan  <http://orcid.org/0000-0003-1348-5310>

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