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Chapter

Dependence of Surface Tension and Viscosity on Temperature in Multicomponent Alloys

Ivaldo Leão Ferreira, José Adilson de Castro and Amauri Garcia

Abstract

Viscosity modeling for pure metals and alloys is widely studied, and many solutions for dependence of viscosity on temperature can be found in the literature for pure metals and alloys. Many of these depend on experimental data for pre-exponential and exponential coefficients. Two key models include: (i) Kaptay model for pure metals, which is completely independent of experimental data and depends only on general constants A and B for a large set of pure metals with few exceptions and (ii) Kaptay viscosity model for liquid alloys derived on the same principles, a temperature-dependent viscosity only as a function of thermophysical properties of the alloy components. In the case of surface tension, the main available models are divided into four groups: Butler formulation-based models, density-functional models, semi-empirical models, and thermodynamic geometric models. Considering the absence of adequate models for surface tension, in this work, two equations relating surface tension and viscosity for pure metals are analyzed as a function of temperature. Regarding the Egry surface tension-viscosity relation for pure metals, a new relation equation for multicomponent alloys is proposed. By applying the proposed equation, the surface tension is calculated and plotted as a function of temperature for ternary and quaternary aluminum alloys.

Keywords: viscosity, surface tension, thermophysical properties, multicomponent alloys, surface tension-viscosity correlation equation

1. Introduction

Surface tension is a phenomenon that occurs whenever a liquid is in contact with other liquids or even gases; then, an interface is established and it acts like a stretched elastic membrane. A surface is called wet if the contact angle is less than 90° and nonwet otherwise. A substrate, like dust or pollution, can contribute energetically to the membrane, decreasing its contact angle. Another feature is the magnitude of the surface tension σ $(N \cdot m^{-1})$. An important effect is the creation of curved meniscus, leading to capillary rise or depression. Viscosity, on the other hand, is a measure of the certain fluid's resistance to flow due to its internal friction. A high viscosity fluid has a trend to resist its motion, such as engine oil, while a fluid with low viscosity flows easily, such as water. Viscosity is a function of fluid's shear

stress and its velocity gradient, and its magnitude η is expressed in $(Pa \cdot s)$. Both viscosity and surface tension are thermodynamic properties of a fluid, and, consequently, can be derived by means of thermodynamic relations.

Many solutions can be found in the literature concerning the viscosity dependence on temperature for pure metals [1–7] and for alloys [8–15]. Budai et al. have reviewed the existing models used to predict dependence of viscosity on temperature of alloys, for cases where the viscosities of pure components are already known [10, 15–18] and those that are independent of experimental data [19]. Budai et al. extended the Kaptay unified equation [10] for the viscosity of alloys, which has been named BBK model [20]. The BKK model was shown to fail in the prediction of viscosity for alloy systems with components that melt congruently [21].

Solutions for surface tension as a function of temperature are generally based on: Butler formulation [22–25]; statistical thermodynamics surface density-functional theory [26–28]; semi-empirical thermodynamic model [29]; and thermodynamic models [30, 31]. All these models are normally specific for certain binary or ternary alloy systems, or they are general but considerably difficult to apply.

In 1992, Egry [32] derived a relation between surface tension and viscosity deduced from statistical mechanics for the melting temperature, based upon the expressions of Fowler [26] for surface tension and Born and Green [33] for viscosity. Both expressions are expressed as integrals over the product of interatomic forces and the pair distribution functions. The author extended this relation to a finite temperature range by using data available in the literature [34].

In this work, by using a straightforward solution for viscosity for molten pure metals [7] and alloys [10], a comparison between numerical simulations and experimental data for the surface tension and viscosity of pure liquid metals and liquid alloys is provided, in order to validate Egry's relation for pure molten metals [34]. An extension of this relation is derived for multicomponent alloys. The surface tension is calculated and plotted against temperature for ternary and quaternary aluminum alloys.

2. Modeling

The modeling section is divided into models dealing with the viscosity of pure metals and multicomponent alloys, and with the surface tension-viscosity relation equations for pure liquid metals and alloys.

2.1 Model for the viscosity of pure liquid metals

Kaptay [7] derived a unified equation for the viscosity of pure liquid metals as a function of temperature, which encompasses the activation energy and the free volume concept. Based on the Andrade's equation [35] as a starting approach, the activation energy concept has been incorporated. By combining again with Andrade's formulation with free volume concept, an equation for the dependence of viscosity of pure metals on temperature has been derived. As this equation obeys both concepts, the authors named it as a unified equation for the viscosity of pure metals. The derived equation for the viscosity of pure metals as a function of temperature is given by,

$$\eta_i = A \frac{M_i^{1/2} \cdot T^{1/2}}{V_i^{2/3}} \cdot \exp \left(B \cdot \frac{T_{m,i}}{T} \right) \tag{1}$$

where η_i is the viscosity of the liquid metal i, A and B are temperature-independent semi-empirical parameters being approximately identical for all liquid

metals, M_i ($kg \cdot mol^{-1}$) is the atomic weight of the metal, V_i is the molar volume ($m^3 \cdot mol^{-1}$), $T_{m,i}$ is the melting temperature of the pure liquid metal i (K), and T is the temperature above the melting point (K).

2.2 Model for the viscosity of multicomponent alloys

An Arrhenius-type viscosity equation can be extended to deal with viscosity of multicomponent alloy, by applying Redlich-Kister polynomial to excess viscosity,

$$\Delta\eta^E = \sum_i \sum_{j>i} x_i \cdot x_j \sum_{k=0}^{m} A_{i,j}^k (x_i - x_j)^k \tag{2}$$

where x_i and x_j are the molar contents of the solute compounds "i" and "j," respectively. $A_{i,j}^k$ are the polynomial parameters related to a binary "ij" system. The ideal viscosity term can be expressed as

$$\eta^{ideal}(T, x_i, x_j) = \sum_{i=1}^{3} x_i \cdot \eta_0^i \exp\left(\frac{E_A^i}{\overline{R} \cdot T}\right) \tag{3}$$

where η_0 is the pre-exponential factor independent of the temperature, interpreted as an asymptotic viscosity at very high temperature, E_A^i is the activation energy of viscous flow of the component i. By combining Eqs. (2) and (3), we obtain,

$$\eta(T, x_i, x_j) = \eta^{ideal}(T, x_i, x_j) + \Delta\eta^E \tag{4}$$

then, we have,

$$\eta(T, x_i, x_j, \cdots, x_n) = \sum_{i=1}^{3} x_i \cdot \eta_0^i \exp\left(\frac{E_A^i}{\overline{R} \cdot T}\right) + \sum_i \sum_{j>i} x_i \cdot x_j \sum_{k=0}^{m} A_{i,j}^k (x_i - x_j)^k \tag{5}$$

where $A_{i,j}^k$ are parameters for the viscosity of binary systems, according to Zhang et al. [36].

Kaptay [10], based on the Seetharaman-Du Sichen equation, regarding the theoretical relationship between the cohesion energy of the alloy and the activation of viscous flow, proposed the following equation:

$$\eta(T, x_i, x_j, \cdots, x_n) = \frac{h \cdot N_{Av}}{\sum_i x_i \cdot V_i + \Delta V^E} \cdot \exp\left[\frac{\sum_i x_i \cdot \Delta G_i^* - \alpha \cdot \Delta H}{\overline{R} \cdot T}\right] \tag{6}$$

where h is the Planck constant ($6.626 \times 10^{-34}\, J.s$), N_{Av} is the Avogadro number (mol^{-1}), ΔG_i^* is the Gibbs energy of activation of the viscous flow ($J \cdot mol^{-1}$), defined as

$$\Delta G_i^* = \overline{R} \cdot T \cdot \ln\left[\frac{\eta_i \cdot M_i}{h \cdot N_{Av} \cdot \rho_i}\right] \tag{7}$$

ΔV^E is the excess molar volume on alloy formation ($m^3 \cdot mol^{-1}$) and α is a ratio of a two properties ratios related to the melting temperature. The first ratio is between the measured activation energy and the melting point of pure liquid metals ($38.4 \pm 2.7\, J \cdot mol^{-1} \cdot K^{-1}$), and the second ratio is the cohesion energy of pure liquid metals and the melting points ($248 \pm 17\, J \cdot mol^{-1} \cdot K^{-1}$), providing $\alpha \cong 0.155 \pm 0.015$ [10].

2.3 Relation between surface tension and viscosity for pure liquid metals

Egry [32] derived a relation between surface tension and viscosity deduced from statistical mechanics for a finite temperature range, based upon the expressions of Fowler [26] for surface tension (σ) and Born and Green [33] for viscosity. Both expressions are expressed as integrals over the product of interatomic forces and the pair distribution functions. The Fowler expression is

$$\sigma = \frac{\pi\, n^2}{8} \int_0^\infty dR \cdot R^4 \, \frac{d\varphi(R)}{dR} \cdot g(R) \tag{8}$$

where n is the particle number density, φ is the pair potential, and $g(R)$ is the correlation function. All these functions depend on the temperature.

In a very similar way, Born and Green [33] derived an expression for the viscosity (η) of a fluid using a kinetic theory, which can be expressed as

$$\eta = \sqrt{\frac{m}{k\, T}} \cdot \frac{2\pi\, n^2}{15} \int_0^\infty dR \cdot R^4 \, \frac{d\varphi(R)}{dR} \cdot g(R) \tag{9}$$

where m is the atomic mass, k is the Boltzmann's constant, and T is the absolute temperature. As the integral terms of both integrals cancel each other, Egry [34] deduced the following relationship between density and viscosity for pure metals for a finite temperature above the melting point:

$$\frac{\sigma_i}{\eta_i} = \frac{15}{16} \sqrt{\frac{k\, T}{m}} \tag{10}$$

where σ_i is the surface tension in $(N.m^{-1})$, and η_i is the viscosity in $(Pa.s)$ of a pure liquid metal i.

In 2005, Kaptay [7] derived a surface tension-viscosity relation, as

$$\frac{\sigma_i}{\eta_i} = \frac{0.182 \cdot (211 + C_{p,i}) \cdot T_{m,i} - (2 + 0.182 \cdot C_{p,i}) \cdot T}{1.61 \cdot M_i^{1/2} \cdot \exp(2.34 \cdot T_{m,i}/T)} \tag{11}$$

where $C_{p,i}$ is the heat capacity of the pure liquid metal i in $(J.mol^{-1}K^{-1})$.

3. A surface tension and viscosity relation model for multicomponent alloys

Based on the formula originally proposed by Egry [34], a new formula is derived for the relation between surface tension and viscosity for multicomponent alloys, that is,

$$\frac{\sigma_a}{\eta_a} = \frac{15}{16} \sqrt{N_{av}\, k\, T} \cdot \frac{1}{\sqrt{\sum_{i=1}^n x_i \cdot M_i}} \tag{12}$$

where σ_a is the alloy surface tension in $(N.m^{-1})$, η_a is the alloy viscosity in $(Pa.s)$, x_i is the molar fraction of the i alloy component, n is the total number of components of the alloy, and M_i is the molar weight of the alloy component i.

Substituting Eq. (12) into Eq. (6), we get,

$$\sigma_a(T, x_i, x_j, \cdots, x_n) = \frac{15}{16} \cdot \frac{h \cdot N_{Av}}{\sum_i x_i \cdot V_i + \Delta V^E} \cdot \sqrt{\frac{\overline{R} \cdot T}{\sum_{i=1}^{n} x_i \cdot M_i}} \cdot \exp\left[\frac{\sum_i x_i \cdot \Delta G_i^* - \alpha \cdot \Delta H}{\overline{R} \cdot T}\right]$$

(13)

4. Results and discussion

Figure 1 presents the viscosity of pure aluminum as a function of temperature, simulated by Kaptay [7] theoretical model compared to experimental data [37]. The model fits the experimental scatter well, but a correction in the melting temperature for pure aluminum was carried out according to the model description provided in [7], although, for the case of Al, no correction was applied by the author [7]. **Figure 2** shows the evolution of viscosity of molten Cu as a function of temperature, where the theoretical model is compared to experimental results [37], where a deviation is noticed for temperatures close to the melting temperature.

In **Figure 3**, the viscosity of pure molten silicon is plotted against temperature. As mentioned, the melting temperature of Si has been assumed as $T_{m\ corr,\ Si} \cong$ 596.85 K [22], which is less than the correction of melting temperature of pure Si proposed by Kaptay [7], that is, $T_{m\ corr,\ Si} \cong$ 870 K. This correction was carried out by the author for some pure molten metals, so that the model could also fit the experimental scatter of these elements. As can be observed, a correction in the melting temperature was needed not only for Si, Ge, Sb, and Bi but also for pure molten Al, based on the same set of experimental data.

In **Figure 4**, it can be seen that Kaptay's model [7] well the experimental data well for the viscosity of molten magnesium as a function of temperature [38–40].

In **Figures 5–8**, the surface tension for pure Al, Cu, Si, and Mg is depicted as a function of temperature, calculated from surface tension-viscosity relations according to Egry [34] and Kaptay [7] formulations. For all cases, except for Si in **Figure 7**, the surface tension exhibits a trend to decrease as the temperature increases. It also can be noticed that for all cases, they diverge close to the melting point. For high temperatures, both models yield very close results. The best agreement observed between the two models is that of pure Mg, as shown in **Figure 8**.

Figures 9–12 show the evolution of viscosity as a function of temperature for all examined alloys, where it can be seen that the two models generally exhibit similar
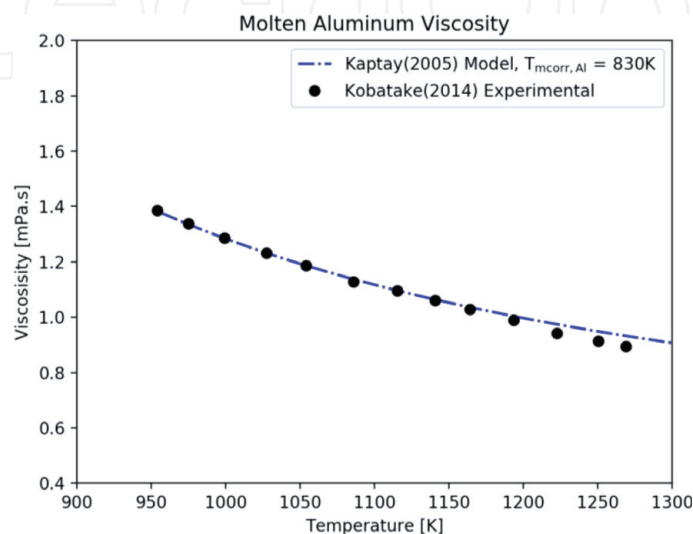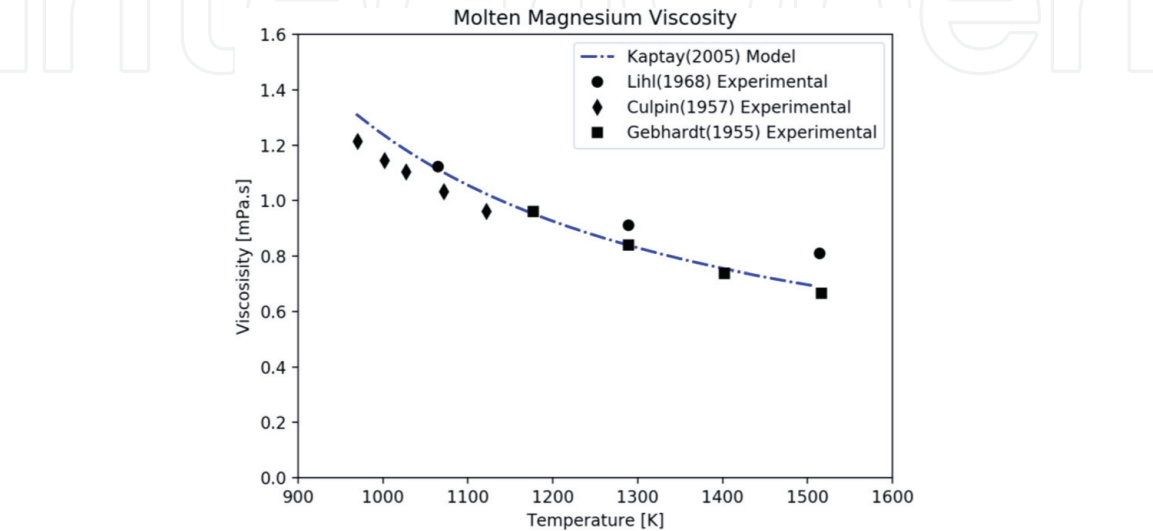

Figure 1.
Calculated viscosities of the pure molten Al as a function of temperature compared to experimental data [37].
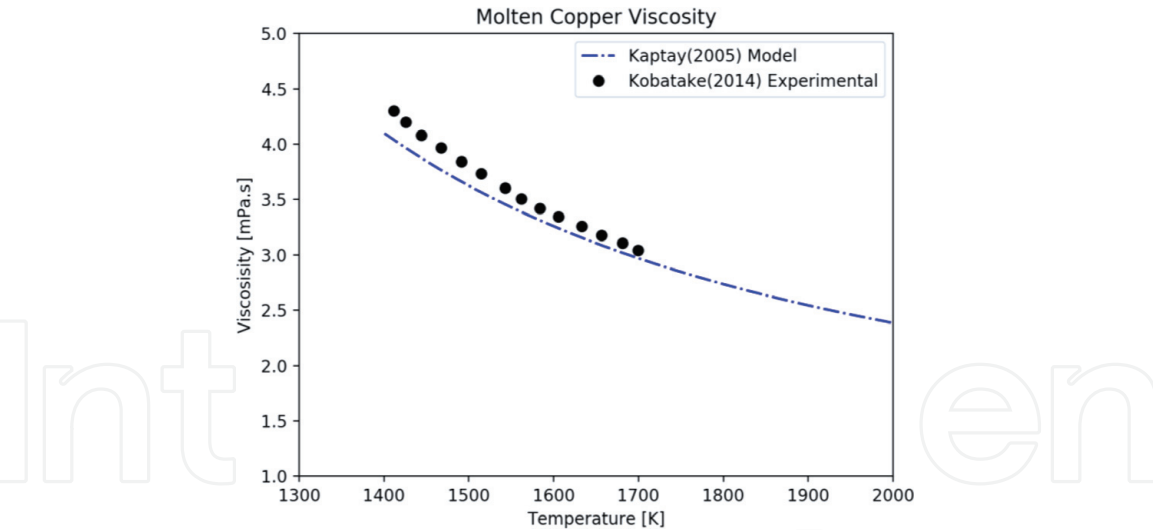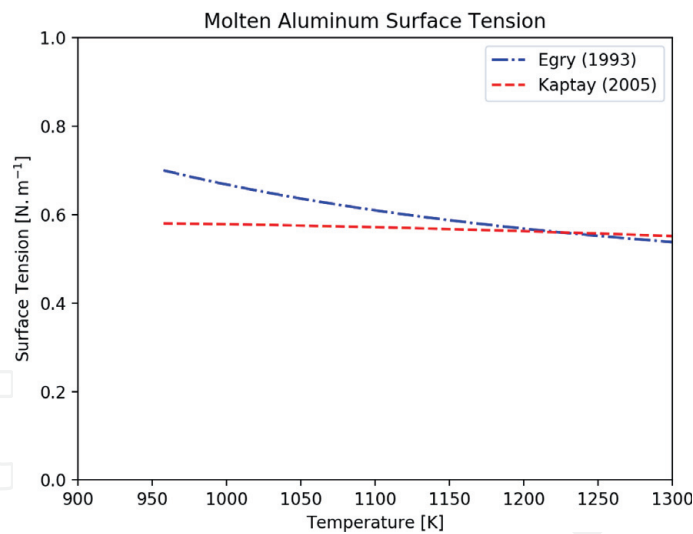
Figure 2.
*Calculated viscosities of the pure molten Cu as a function of temperature compared to experimental data [37].*
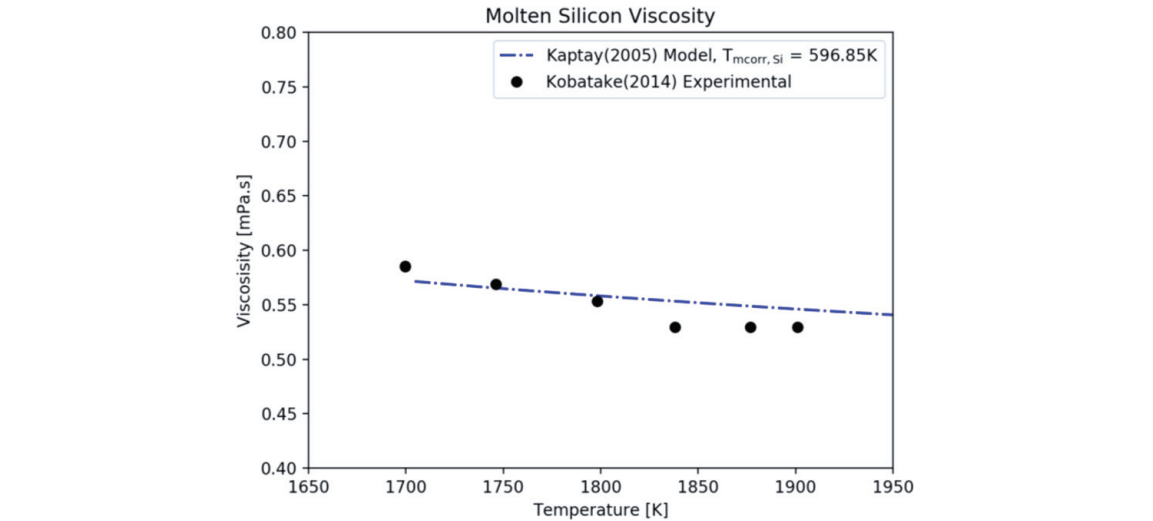
Figure 3.
Calculated viscosities of the pure molten Si as a function of temperature compared to the experimental data [37].
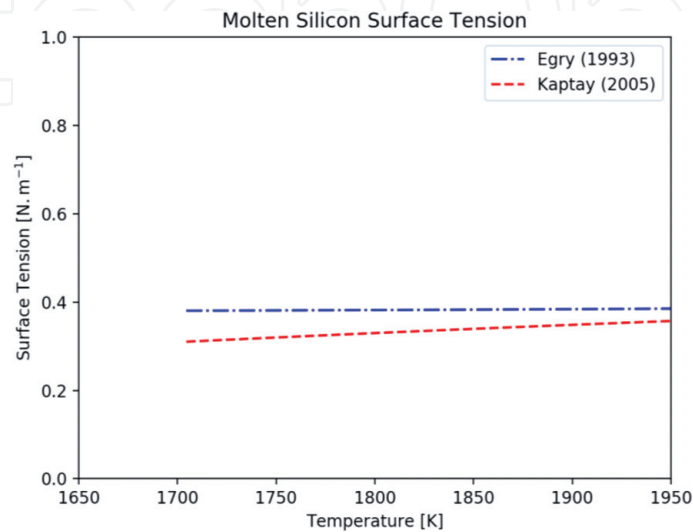

Figure 4.
Calculated viscosities of the pure molten Mg as a function of temperature compared to the experimental data.

Figure 5.
Comparison between surface tension of pure molten Al as a function of temperature provided by both Egry's and Kaptay's surface tension-viscosity relation models.
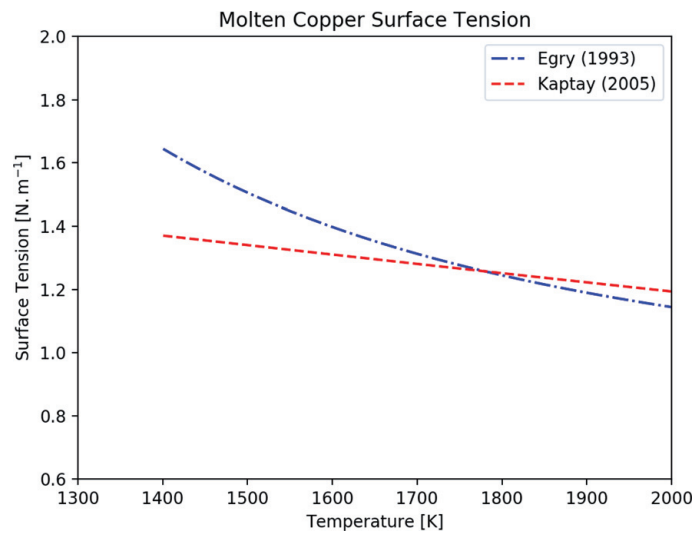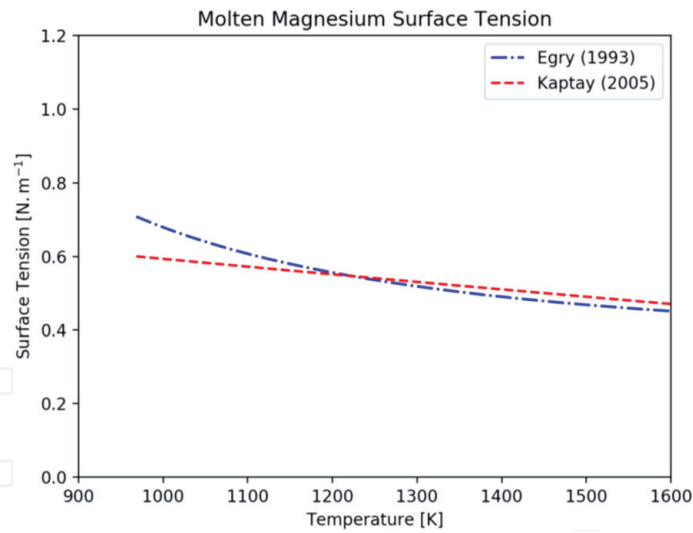

Figure 6.
Comparison between surface tension of pure molten Cu as a function of temperature provided by both Egry's and Kaptay's surface tension-viscosity relation models.
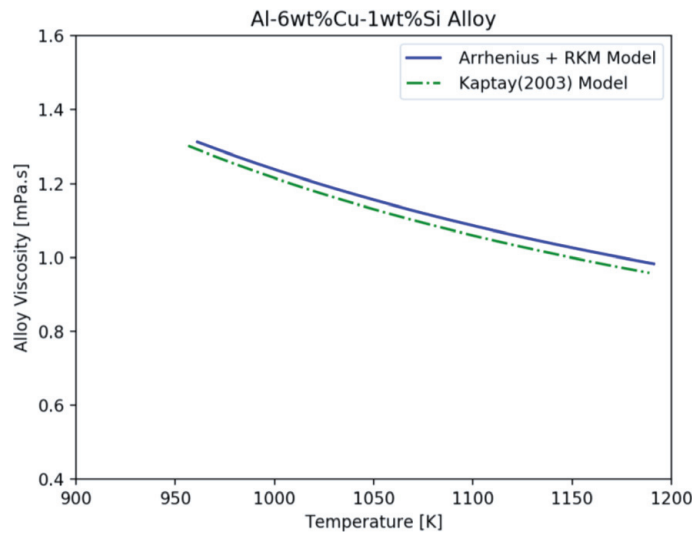

Figure 7.
Comparison between surface tension of pure molten Si as a function of temperature provided by both Egry's and Kaptay's surface tension-viscosity relation models.
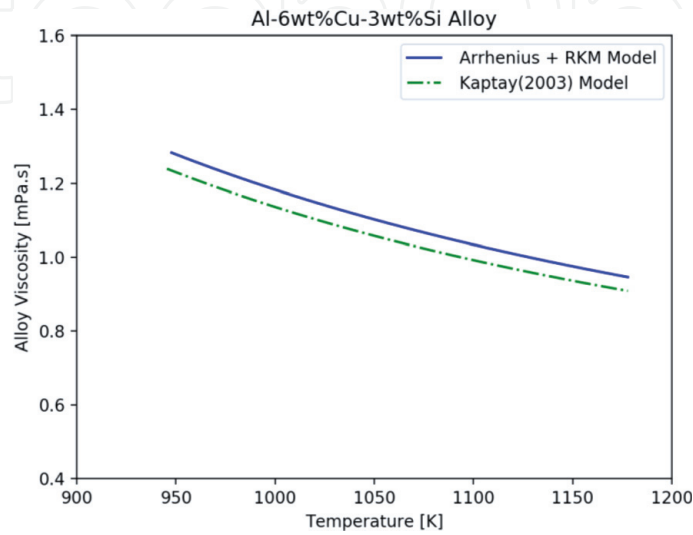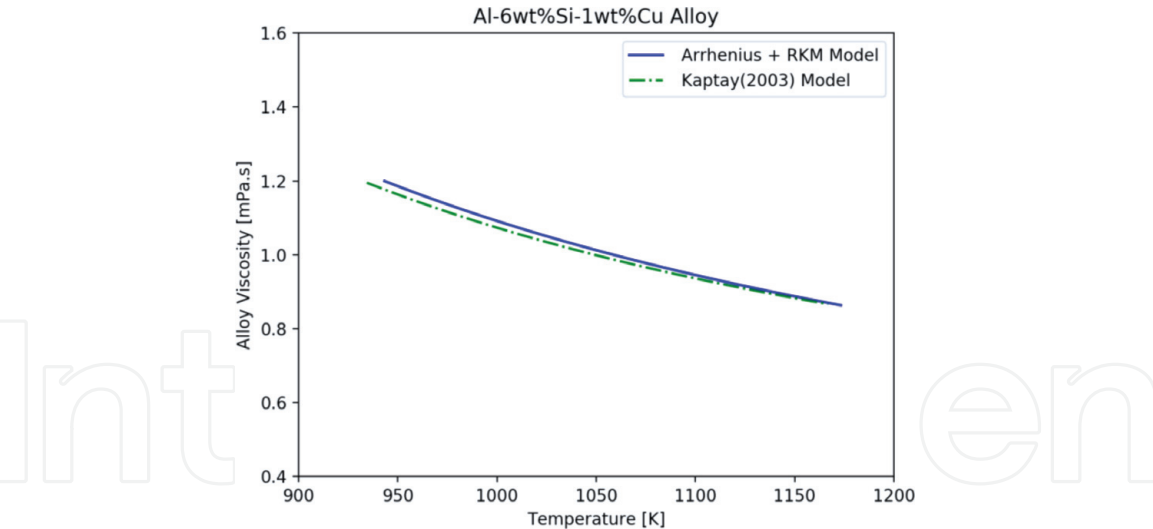
Figure 8.
Comparison between surface tension of pure molten Mg as a function of temperature provided by both Egry's and Kaptay's surface tension-viscosity relation models.
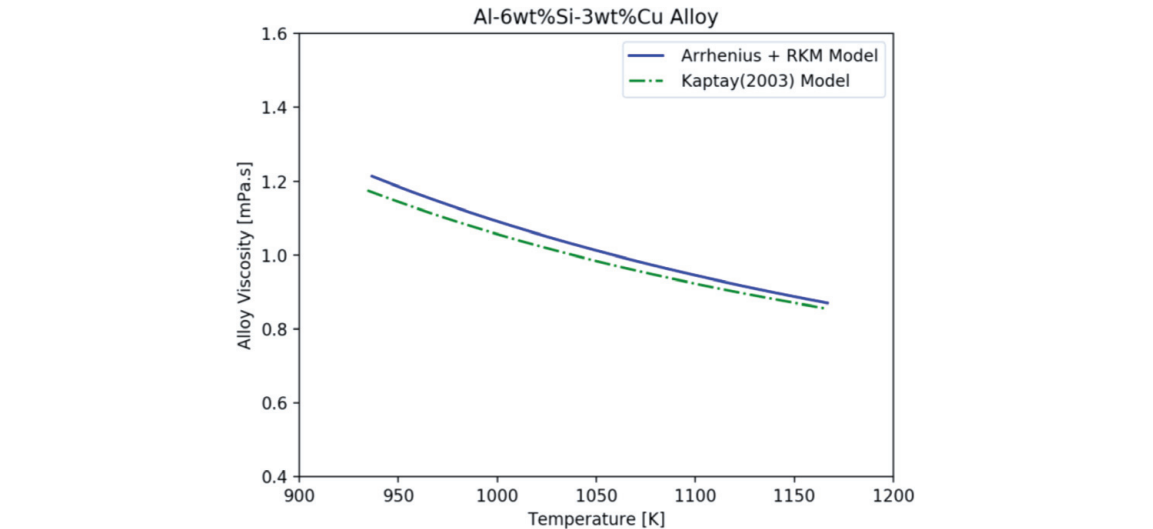
Figure 9.
Simulations of viscosity of Al-6wt%Cu-1wt%Si as function of temperature: Arrhenius-type equation and Kaptay model.
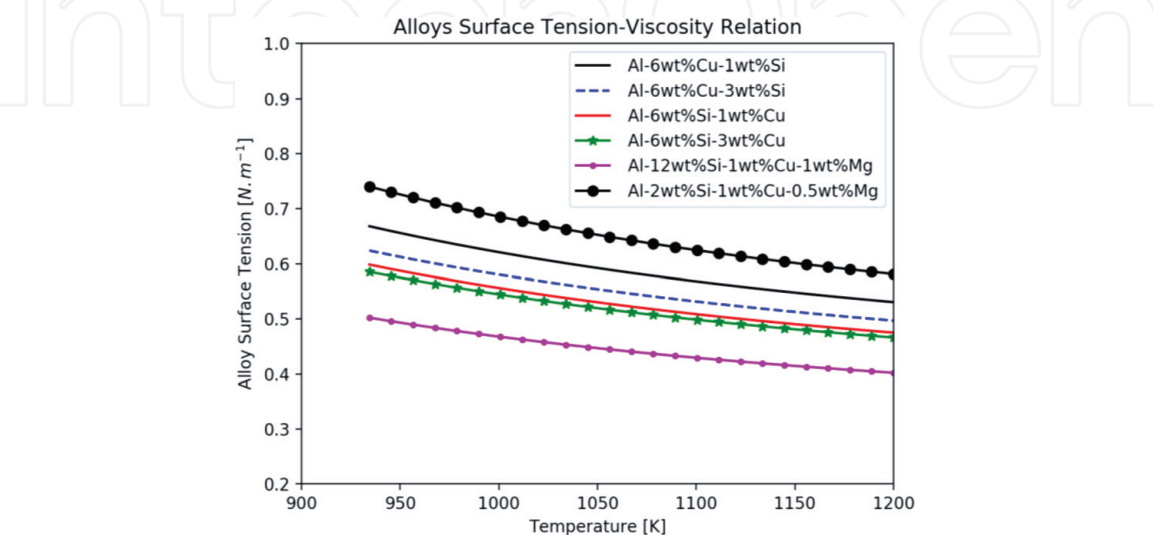
Figure 10.
Simulations of viscosity of Al-6wt%Cu-3wt%Si as function of temperature: Arrhenius-type equation and Kaptay model.

Figure 11.
Simulations of viscosity of Al-6wt%Si-3wt%Cu as function of temperature: Arrhenius-type equation and Kaptay model.

Figure 12.
Simulations of viscosity of Al-6wt%Si-3wt%Cu as function of temperature: Arrhenius-type equation and Kaptay model.

Figure 13.
Application of derived surface tension-viscosity relation for Al-Cu-Si ternary alloys and quaternary Al-Cu-Si-Mg alloys as function of temperature.

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results. The greatest deviation between the models can be observed for the Al-6wt% Cu-3wt%Si alloy, **Figure 10**; but even in this case, a relatively good agreement can be considered. This may be related to the non-ideal term of viscosity of the Redlich-Kister equation, whose coefficients do not depend on the alloy composition but depend on the temperature [36].

Figure 13 shows the application of the derived surface tension relation equation for alloys, Eq. (13), to ternary Al-Cu-Si and quaternary Al-Cu-Si-Mg alloys. As can be noticed, the increase in the alloy Si content decreases the surface tension. The lowest surface tension profile is associated with the alloy having the highest Si content, that is, for the Al-12wt%Si-1wt%Cu-1wt%Mg quaternary alloy.

5. Conclusion

A solution for viscosity of alloys has been derived based on an extension of Egry's surface tension-viscosity relation equation for pure metals [34] and Kaptay's [10] unified solution for viscosity of multicomponent alloys. Kaptay and Egry's surface tension-viscosity relations were plotted against temperature. It was shown that for the pure metals analyzed in the present study, for lower temperatures, both relations deviated from each other. However, for higher temperatures, a better agreement has been achieved. The derived solution for surface tension was plotted against temperature for ternary Al-Cu-Si and Al-Cu-Si-Mg alloys. It was shown that with increasing Si alloy content, the surface tension of Al-based alloys decreases, which is in agreement with the casting practice of Aluminum-based alloys.

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