Viscosity of Al–Cu liquid alloys: measurement and thermodynamic description

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Abstract In the present work a high temperature oscillating cup viscometer has been used to measure the viscosities of liquid binary Al–Cu alloys. The dependence of viscosity on temperature is well described by the Arrhenius law. For constant temperature, the viscosity as a function of copper concentration exhibits a maximum at a mole fraction $x_{Cu} = 0.7$. This might be due to a pronounced chemical short range order in the liquid phase at this composition. As the comparison of existing phenomenological models describing viscosity as a function of composition to the experimental data is unsatisfactory, a new model for the viscosity has been developed within this work based only on a few assumptions and using the enthalpy of mixing as input parameter which is easily accessible. The agreement between model calculation and experimental data is excellent.

Introduction

Viscosity is an outstanding thermophysical property which is of great interest for the study of solidification of industrial cast alloys and for the simulation of solidification processes involving fluid flow. Among the cast alloys, the binary Al–Cu system has found many applications, mainly as light weight alloys in the automotive and aeronautic industry. On the one hand, it is desirable to create a highly accurate and reliable set of thermophysical property data of the binary Al–Cu system which, on the other hand, is the prerequisite to correctly predict thermophysical properties of ternary or higher order systems containing Al and Cu. One example is the ternary Al–Cu–Ag system, which lead free solders are based on. Densities and viscosities of alloys in this system have been measured by Brillo et al. [1]. Surface tensions have been determined in electromagnetic levitation by [2–5] using the oscillating drop technique and densities and excess volumes [6, 7] again in electromagnetic levitation by a shadowgraph technique.

Jones and Bartlett [8] measured the viscosities of Al–Cu binary alloys up to a maximum Cu concentration of $x_{Cu} = 0.173$, corresponding to the composition of the first eutectic in the phase diagram. Gebhardt et al. [9] also used the oscillating cup technique to determine viscosities of Al–Cu alloys in the Al-rich corner up to a maximum Cu content of $x_{Cu} = 0.05$, Lihl et al. [10] up to $x_{Cu} = 0.215$. Two of these measurements [8, 9] have been performed at a time when the Roscoe [11] equation has not yet been developed.

Due to the rather high melting temperature of Cu, a comprehensive study of Cu-rich alloys has not been possible in the past. The maximum temperature for the viscosity measurements of Jones and Bartlett [8] is restricted to 1123 K. For Gebhardt et al. [9], the maximum temperature for their viscosity measurements is 1273 K, and the temperature-dependent viscosities of Al–Cu reported by Lihl et al. [10] are limited to a maximum temperature of 1173 K. Gebhardt et al. [9] determined the temperature dependence of the viscosity between 1173 K and the melting point in steps of 100 K, resulting in four data points per alloy. The absolute values for viscosity in

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dependence of temperature of Jones and Bartlett [8] differ quite largely from those of Gebhardt et al. [9].

More recent measurements have been carried out by Friedrichs et al. [12], Plevachuk et al. [13], and Konstantinova and Popel [14]. Plevachuk et al. [13] determined viscosities of hypoeutectic Al–Cu alloys using an oscillating cup viscometer and Konstantinova and Popel [14] investigated the system up to $x_{Cu} = 0.5$. The maximum temperature of both results was about 1250 K. In the work of Friedrichs et al. [12], a gas bubble viscometer has been employed for the measurement of viscosity, density, and surface tension. In this method a spherical bubble shape is assumed and the viscosity is obtained from Stokes' law. Although they reported viscosities for temperatures of up to 1600 K, only Al-rich alloys close to the eutectic composition have been investigated. In summary, there is a considerable lack of viscosity data on the Cu-rich side of the binary Al–Cu system. Moreover, the reported viscosities are not consistent with each other.

On the other hand, with our high temperature oscillating cup viscometer [15] we can reach a maximum temperature of 2200 K while a highly pure and chemically inert atmosphere is maintained.

In addition to the experimental facts, also several models can be found in literature describing the dependence of viscosity on composition. An overview on them is given, for instance, in Ref. [16]. To some extent, they are all based on thermodynamic parameters and predict the experimental data of binary alloys with varying accuracy [17].

The model of Moelwyn-Hughes [18] relates the deviation of the viscosity from ideal behavior to the enthalpy of mixing of the two components A, B

$$\eta = (x_A\eta_A + x_B\eta_B)\left(1 - 2\frac{\Delta H_{mix}}{RT}\right). \tag{1}$$

In Eq. (1) η_A and η_B are the viscosities and x_A and x_B the mole fractions of components A, B in the alloy, ΔH_{mix} is the enthalpy of mixing, R the universal gas constant, and T the temperature.

Kozlov et al. [19] considered atomic vibration frequencies to describe the Gibbs energy. For an alloy consisting of N components, their model results in

$$\ln \eta = \sum_{(i=1)}^{N} x_i\ln \eta_i - \frac{\Delta H_{mix}}{3RT}. \tag{2}$$

η is the viscosity of the alloy, η_i are the viscosities and x_i the mole fractions of the pure components.

Seetharaman and Sichen [20] started from an Arrhenius-type approach

$$\eta = \frac{hN_A}{v} \cdot \exp\left(\frac{\Delta G^*}{RT}\right) \tag{3}$$

where h is Planck's constant, N_A Avogadro's number, v the molar volume, and ΔG^* the Gibbs activation energy of the viscous flow. ΔG^* can be expressed by the Gibbs activation energies of the pure components ΔG_i^* and the Gibbs energy of mixing ΔG_{mix}. The model of Seetharaman and Sichen [20] yields the following equation for a binary alloy consisting of the components A and B:

$$\eta = \frac{hN_A}{v} \cdot \exp\left(\frac{x_A\Delta G_A^* + x_B\Delta G_B^* + \Delta G_{mix} + 3RTx_Ax_B}{RT}\right) \tag{4}$$

A similar expression is derived by Kaptay [21], who introduces the enthalpy of mixing ΔH_{mix} multiplied with a semi-empirical factor $\alpha = 0.155$, which has been adjusted to a number of certain binary alloys.

$$\eta = \frac{hN_A}{v} \cdot \exp\left(\frac{x_A\Delta G_A^* + x_B\Delta G_B^* - (0.155 \pm 0.015) \times \Delta H_{mix}}{RT}\right) \tag{5}$$

In this equation, the molar volume v is given by

$$v = \sum_i x_i v_i + \Delta^E v \tag{6}$$

Here, v_i is the molar volume of component i and $\Delta^E v$ the molar excess volume.

Hirai [22] correlates the activation energy with the liquidus temperature T_L:

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

M is the molar mass in kg mol^{-1}, ρ the density in kg m^{-3}. In this equation, all quantities needed for calculation are usually known. In the following, we present the model we developed to describe our experimental data.

Experimental

All measurements have been carried out in a high temperature oscillating cup viscometer. Details about the experimental setup are described in literature [15]. At the beginning of an experiment, the solid sample is contained in a cylindrical crucible made of high temperature resistant material, alumina in this case. The crucible is encapsulated in an outer graphite container, attached to the torsion wire and placed inside the high temperature furnace. The furnace is part of a high vacuum system, which is first evacuated to a pressure of $<2 \times 10^{-6}$ mbar and then backfilled with 400 mbar argon of high purity, 99.999 %. The sample

is molten under argon atmosphere. Torsional oscillations of the suspended crucible are excited by an electric motor. The time dependent angular displacement of the oscillation system is amplified by a laser beam, reflected from a mirror, placed in the rotation axis. The reflected light impinges onto a position sensitive detector (PSD) which is read out by dedicated software. This software records the time dependent oscillation and calculates the viscosity by solving the Roscoe equation [23] for the motion of the liquid numerically. The corrected version of the Roscoe equation was used without the known misprint in the original paper [11].

A pyrometer, calibrated at the liquidus temperatures of several pure metals (Al, Cu, Ni, Co, Fe) records the temperature during the experimental run. The temperature is slowly decreased in steps of 1 K/min during data acquisition. One experimental run yields several hundred data points over a wide temperature range. They generally follow an Arrhenius' law for the viscosity:

$$\eta = \eta_\infty \cdot \exp\left(\frac{E_A}{RT}\right). \tag{8}$$

Here η_∞ is the pre-exponential factor, which can be interpreted as an asymptotic viscosity for very high temperatures. E_A is the activation energy of the viscous flow, R the universal gas constant, and T the temperature. From the Arrhenius fits for alloys of different composition, the dependence of viscosity upon composition can easily be derived by calculating isotherms for several constant temperatures.

Starting materials for alloy preparation were the pure elements Cu (nuggets, >99.99 %, Umicore Belgium) and Al (granules, 99.99 %, ChemPur Germany). Prior to sample preparation, the required weights of Cu and Al were calculated corresponding to the desired composition. The calculated masses of the elements were cut, polished, rinsed with water and isopropanol. Cu was additionally cleaned with concentrated acetic acid (50 %) to remove surface oxides and rinsed with water and isopropanol for a second time. Finally the prepared amounts of Cu and Al were put into a cup made from alumina (Haldenwanger, Germany) which had also been cleaned with isopropanol before. For Cu-rich alloys, cups of 50 mm height and 16.5 mm in diameter were used. Because of the considerably lower density of Al compared to Cu, for Al-rich alloys larger cups with 26 mm in diameter and the same height had to be employed. This procedure is justified as for the Al-rich melts the viscosity is comparatively low. The total masses of the samples were kept in a range from 21 to 28 g, the deviation from the nominal composition was lower than 0.1 %. The cup was covered by a lid and enclosed into an outer graphite container which was attached to the oscillation system of the viscometer. Alloys

were prepared "in situ" by heating the mixed pieces of Cu and Al in the furnace to 1873 K. At constant temperature the liquid mixture was allowed for homogenization until the recorded viscosity value was constant for at least half an hour. Then, the temperature was decreased continuously by 1 K/min while measuring the viscosity in dependence of temperature until solidification of the sample was observed.

Results and discussion

Viscosity as a function of temperature and composition has been measured for the binary Al–Cu system covering the whole composition range.

Viscosity data for the pure components Al and Cu have been taken from literature [17, 24]. The value for the viscosity of pure Al, quoted in ref 24 is slightly higher than the recommended value given by Assael et al. [25]. For consistency with our previous publication [1], we have chosen to use the value of ref 24. For $Al_{0.8}Cu_{0.2}$ existing data have been included measured earlier by Brillo et al. [26]. Figure 1a shows the results for viscosity as a function of temperature compared to the pure components.

A broad temperature range from 830 to 1600 K is covered by our measurements. This corresponds to a change in the inverse temperature, T^{-1}, by a factor of nearly two. The scatter of the data is approximately ±10 %. For Al-based melts, this can be regarded as low. With respect to the dependence of the viscosity on composition, two facts become obvious from Fig. 1a: Firstly, pure Al shows the lowest viscosity compared to the alloys measured in the present work. Secondly, for Cu mole fractions in the range between $x_{Cu} = 0.5$ and 0.9 the absolute values of viscosity are exceeding those of pure Cu. It follows that, for constant temperature, the viscosity exhibits a maximum for a certain composition.

For some compositions ($Al_{0.9}Cu_{0.1}$, $Al_{0.5}Cu_{0.5}$, $Al_{0.7}Cu_{0.3}$) slight deviations from the Arrhenius functions occur in the region of $10^4/T < 6$ K^{-1}. As we could exclude that this was caused by a possible loss of mass, these deviations are unexpected to some extent.

In the case of $Al_{0.4}Cu_{0.6}$ the scatter of the experimental data is slightly larger than for the other measurements. During processing of this specific sample exceptional vibrations occurred in the lab from which the sample could not sufficiently be protected.

From an Arrhenius fit to the experimental data, the energies of activation of the viscous flow E_A and the asymptotic viscosity η_∞ are obtained as functions of composition. The results are compiled in Table 1.

The Arrhenius functions, calculated from the fit parameters are shown in Fig. 1b where the viscosity is plotted versus temperature.
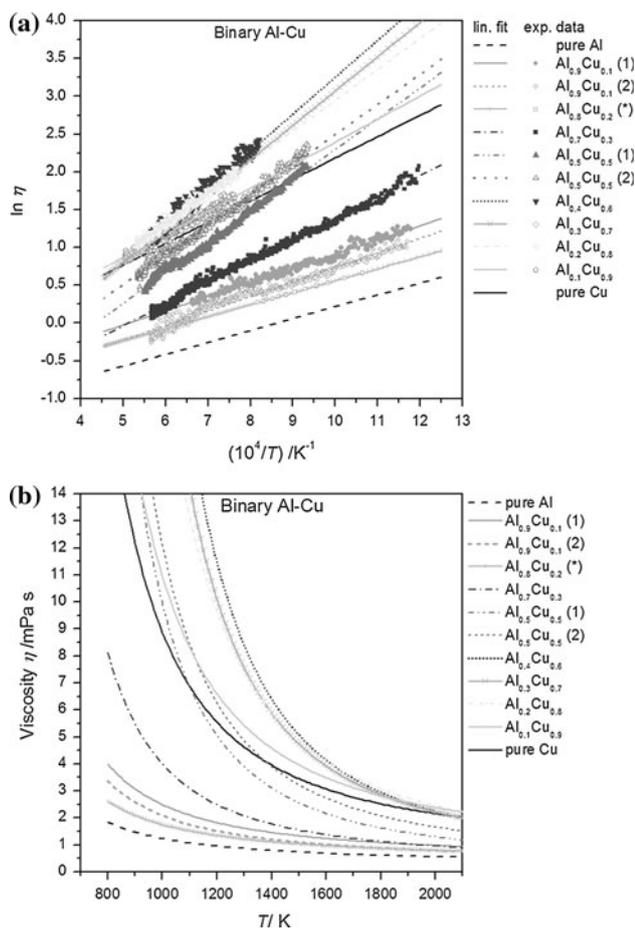
Fig. 1 a Measured logarithmic viscosity $\ln \eta$ as a function of the inverse temperature $10^4/T$ for several binary Al–Cu alloys in comparison to the pure components Al and Cu [17, 24]. Measurements of samples with the same composition are denoted with numbers, $Al_{0.8}Cu_{0.2}$ (*) has been measured before by Brillo et al. [26]. **b** Fitted Arrhenius functions for viscosity in dependence of temperature compared to the pure components Al and Cu [17, 24]. For better clarity, the experimental data points are not shown. Measurements of samples with the same composition are denoted with numbers, $Al_{0.8}Cu_{0.2}$ (*) has been measured before by Brillo et al. [26]

Despite the scatter of the data, especially for $Al_{0.4}Cu_{0.6}$, the statistics of the Arrhenius fits are very good due to the large number of data points recorded per experimental run, which is one of the advantages of the used high temperature viscometer. From the Arrhenius functions fitted to the experimental data, the viscosity can be calculated for binary Al–Cu as a function of composition. The isotherms in Fig. 2 show a maximum in viscosity between $x_{Cu} = 0.6$ and 0.8. In the region of the maximum several intermetallic phases in the solid exist as can be seen in the phase diagram of binary Al–Cu (Fig. 3), calculated from the thermodynamic description by Witusiewicz et al. [27].

The same phenomenon has been observed for binary Al–Ni [28], In–Sn and In–Bi alloys [29]. For certain compositions in the Al–Ni system, where solid

intermetallic phases exist in the phase diagram, intensified chemical short range order in the liquid, reminiscent of the compound formation in the solid, is discussed as the reason for an increase in viscosity [30–32].

For Al–Cu, however, it was shown that, in the case of surface tension, compound formation in the liquid can be ignored in the thermodynamic description and the (sub-)regular solution model is sufficient [4]. As will be shown further below the model is sufficiently precise without considering compound formation in the model.

In literature several models can be found, which describe viscosity as a function of composition for binary alloys in dependence of thermophysical properties like ΔH_{mix} or T_L. They are compared to the experimental data in Fig. 2. The required input quantities, such as ΔH_{mix}, ΔG_{mix}, $^E G_{mix}$ and T_L as functions of composition have been calculated from CALPHAD models as described for Al–Cu further below. Densities and molar excess volumes are available from [6]. The model of Moelwyn-Hughes [18] predicts the position of the maximum in viscosity correctly between $x_{Cu} = 0.6$ and 0.8, but overestimates the absolute values considerably. This behavior has also been observed for binary Al–Ni alloys by Kehr et al. [28]. The model of Kozlov et al. [19] also shows a weak maximum at about $x_{Cu} = 0.7$, but underestimates the absolute values in the region from $x_{Cu} = 0.5$–0.8. The curve calculated from the model of Kaptay [21] exhibits a similar shape without a pronounced maximum, predicting even lower viscosities. The same applies for the model of Hirai [22], which correlates the viscosity to the liquidus temperature of the alloy in dependence of composition (liquidus line in the phase diagram). The curve calculated from the model of Seetharaman and Sichen [20] shows a minimum in viscosity instead of the expected maximum.

In summary, the comparison of the viscosity models described in literature to the experimental data of this work (Fig. 2) clearly indicates, that the agreement is not satisfactory so far.

Modeling

Model developed within the present work

As a consequence of the non-satisfactory agreement between the existing models and our data, we pursued the question, if it is possible to describe viscosities by a simple approach, based only on a few assumptions.

One assumption is that the activation energy of viscous flow is generally larger the more attractive the interactions between the atoms are. This is due to the fact that shear flow can only take place if nearest-neighbor-bonds are

Table 1 Activation energies of the viscous flow E_A, and asymptotic viscosities η_∞ for Al–Cu binary alloys, determined from fits of the Arrhenius law Eq. (1) to the experimental data as functions of temperature

x_{Cu}	η_∞ (mPa s)	E_A (kJ mol^{-1})	T_L (K)
0	0.257	13,100	933
0.1	0.305	16,014	873
0.1	0.380	15,640	873
0.2	0.358	13,203	834
0.3	0.233	23,630	867
0.4	–	–	976
0.5	0.228	33,058	1105
0.5	0.169	33,873	1105
0.6	0.195	40,666	1228
0.7	0.229	37,789	1303
0.8	0.316	34,089	1313
0.9	0.517	25,409	1346
1	0.520	23,570	1358

Viscosity data for $x_{Cu} = 0.2$ has been measured by Brillo and co-workers [25]. Data for the pure components are from Refs. [17, 24]

Melting temperatures T_L from CALPHAD calculations of this work, based on Witusiewicz et al. [27]

Fig. 3 Phase diagram of binary Al–Cu calculated with CALPHAD. Phases have been modeled according to the thermodynamic description of Witusiewicz et al. [27]

$$\hat{E}_A^{Alloy} = x_{Al}E_A^{Al} + x_{Cu}E_A^{Cu} - \Delta H_{mix}(x_{Al}) + RT(x_{Al}\ln x_{Al} + x_{Cu}\ln x_{Cu}) \tag{9}$$

where $x_{Al} = 1 - x_{Cu}$ is the mole fraction of Al, and E_A^{Al} and E_A^{Cu} are the activation energies for the pure components Al and Cu, respectively. ΔH_{mix} is the enthalpy of mixing which, for Al–Cu, is obtained from CALPHAD modeling as described in a following section. The last term in Eq. (9) accounts for the entropy of mixing which is approximated as ideal.

Moreover, it has been argued [33] that, in the simplest way, the pre-exponential factor of the alloy, $\hat{\eta}_\infty$, is expressed as a function of the concentrations in the following way, if η_∞^{Al} is the pre-exponential factor of Al and η_∞^{Cu} the one of Cu:

$$\ln \hat{\eta}_\infty = x_{Al}\ln \eta_\infty^{Al} + x_{Cu}\ln \eta_\infty^{Cu} \tag{10}$$

Equations (9) and (10) are inserted into Eq. (8). One must note that in the Arrhenius equation, the activation energy, E_A, must not depend on temperature. Therefore, the last term in Eq. (9) is factored out from the exponent and contributes to the pre-exponential factor η_∞. The following expressions are thus obtained for E_A and η_∞:

$$E_A = x_{Al}E_A^{Al} + x_{Cu}E_A^{Cu} - \Delta H_{mix}(x_{Al}) \tag{11}$$

$$\ln \eta_\infty = x_{Al}\ln(x_{Al}\eta_\infty^{Al}) + x_{Cu}\ln(x_{Cu}\eta_\infty^{Cu}) \tag{12}$$

Thermodynamic phase modeling for binary Al–Cu (CALPHAD)

For model calculations of the viscosity as a function of composition using the models described before, the Gibbs energy of mixing ΔG_{mix}, the enthalpy of mixing ΔH_{mix},
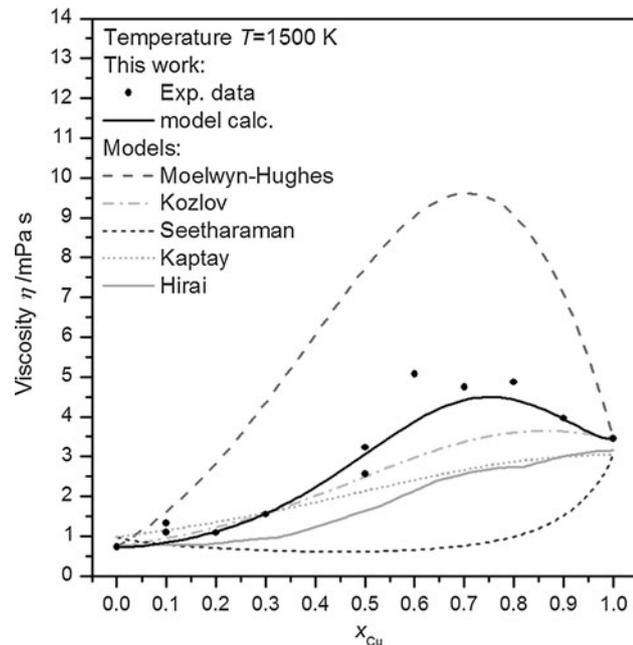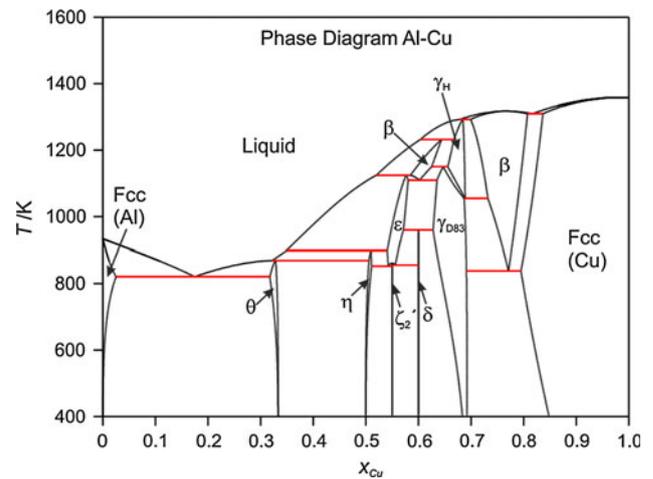

Fig. 2 Comparison of experimental viscosity for Al–Cu as a function of composition to viscosity calculated from several models including the one developed in this work. The results are shown for constant temperature $T = 1500$ K

constantly being broken. Hence, the activation energy of viscous flow in an alloy, $\hat{E}_A^{Alloy}$, can be written (for Al–Cu in this case) as

and the excess Gibbs energy ^EG are needed. A convenient access to these quantities is provided by the CALPHAD method, which has been used in the present work to calculate ΔG_{mix}, ΔH_{mix}, and $^EG_{\mathrm{mix}}$ for the liquid phase of the binary Al–Cu system. The calculations are carried out in dependence of composition and temperature, based on the thermodynamic modeling of Witusiewicz et al. [27]. Also, the liquidus temperatures T_{L} (liquidus line in the phase diagram) used in the Hirai model [22] have been calculated with the CALPHAD approach.

The Gibbs energies of the elements Al and Cu $^0G_i^\varphi$ (i = Al, Cu) in the phase φ as functions of temperature are given by a polynomial of the following form [34]:

$$^OG_i^\varphi = G_i^\varphi - H^{\mathrm{SER}}$$
$$= a_0 + a_1 T + a_2 T \ln T + a_3 T^2 + a_4 T^3 + a_5 T^{-1}$$
$$+ a_6 T^7 + a_7 T^{-9} \tag{13}$$

Reference state is the stable element reference state (SER), i.e., the enthalpy H^{SER} at $T = 298.15$ K and $p = 1$ bar. Parameters a_0–a_7 for Gibbs energy functions of pure components are given by the Scientific Group Thermodata Europe (SGTE) [34].

The phases in the binary Al–Cu system have already been modeled by Saunders [35], and revisited by Witusiewicz et al. [27]. The following phases have been modeled as disordered solutions: Liquid, fcc-Al (α-phase), bcc-A2 (β-phase), and hcp-A3. The molar Gibbs energy $G_{\mathrm{m}}^\varphi(T,x)$ of phase φ in dependence of temperature T and composition is given by Eq. (14):

$$G_{\mathrm{m}}^\varphi(T,x) = x_{\mathrm{A}}{}^OG_{\mathrm{A}}^\varphi(T) + x_{\mathrm{B}}{}^OG_{\mathrm{B}}^\varphi(T) + RT(x_{\mathrm{A}} \ln x_{\mathrm{A}}$$
$$+ x_{\mathrm{B}} \ln x_{\mathrm{B}}) + {}^EG^\varphi \tag{14}$$

$^OG_{\mathrm{A}}^\varphi$ and $^OG_{\mathrm{B}}^\varphi$ are the molar Gibbs energies of the pure components A and B in phase φ, $^EG^\varphi$ is the excess Gibbs energy of phase φ and can be written as a Redlich–Kister polynomial [36]:

$$^EG_{\mathrm{A,B}}^\varphi = x_{\mathrm{A}} x_{\mathrm{B}} \sum_{v=0}^{n} {}^v L_{\mathrm{A,B}}^\varphi (x_{\mathrm{A}} - x_{\mathrm{B}})^v, \tag{15}$$

where $^v L_{\mathrm{A,B}}^\varphi$ are the interaction parameters of order v for the pure components A and B in phase φ. The dependence of $^v L_{\mathrm{A,B}}^\varphi$ on temperature is linear as described by Witusiewicz et al. [27] with $v = 3$ for the order of $^v L_{\mathrm{A,B}}^\varphi$:

$$^v L_{\mathrm{A,B}}^\varphi = a_v^\varphi + b_v^\varphi T \tag{16}$$

The sublattice model introduced by Hillert and Staffansson [37] and extended by Sundman and Ågren [38] offers a convenient method of description for the ordered phases in the binary Al–Cu system, even for the stoichiometric ones. For most phases two sublattices are sufficient: δ-phase

(Al)$_2$:(Cu)$_3$, ζ-phase (Al)$_9$:(Cu)$_{11}$, ε- and η-phase (Al, Cu):(Cu), and θ-phase (Al)$_2$:(Al, Cu). The phases γ (ordered) and γ-H (high temperature γ-phase) have been modeled using three sublattices: (Al)$_4$:(Al, Cu):(Cu)$_8$. The notation of the G functions corresponds to Sundman and Ågren [38], the excess Gibbs energies are written as Redlich–Kister polynomials, compare Eqs. (15) and (16).

Model application

The viscosity calculated from our model for $T = 1500$ K is also shown in Fig. 2, the agreement with the experimental data is excellent. Not only the position of the maximum is reproduced at $x_{\mathrm{Cu}} \approx 0.7$, also the absolute values of viscosity are predicted correctly. For $T = 1500$ K, the calculated viscosities, the experimentally determined ones, and the deviations of both from each other are shown in Table 2.

Furthermore, the activation energy of the viscous flow in dependence of composition can be calculated from our model (Eq. 11). The result is compared to the experimentally determined activation energies (Eq. 8) in Fig. 4.

They compare excellently to the experimental data. The same is true for the asymptotic viscosities η_∞. The comparison between model and experiment is given in Fig. 5.

It should be noted in this place that Eq. (11) depends on the mixing enthalpy, ΔH_{mix}, which, in principle, could have any mathematical form. Hence, it could also be calculated from a model including the effect of compound formation in the liquid phase. Equation (15), however, represents a model for the (sub-)regular solution which appears to be sufficient for the description of the experimental viscosity data. Like for the surface tension [4], concepts about compound formation in the liquid phase do not need to be considered.

Summary

In the present work the viscosities of liquid binary Al–Cu alloys have been measured using a high temperature oscillating cup viscometer. The temperature dependence is described by the Arrhenius law yielding the asymptotic viscosity η_∞ and the energy of activation of the viscous flow E_{A} as fit parameters. The dependence of viscosity upon composition has been analyzed for constant temperature $T = 1500$ K. A maximum of viscosity is found at $x_{\mathrm{Cu}} \approx 0.7$, where the attractive interaction is particularly pronounced. For the description of viscosity as a function of composition, a model has been developed within the present work. It is based on only a few but physically justified assumptions. The model compares excellently to the experimental data, describing viscosity, energy of

Table 2 Viscosities as functions of composition (x_{Cu}) for $T = 1500$ K determined from the experiments (η_{exp}) and from our model (η_{theo})

x_{Cu}	η_{exp} ($T = 1500$ K) (mPa s)	η_{theo} ($T = 1500$ K) (mPa s)	$\left\vert \frac{\eta_{theo} - \eta_{exp}}{\eta_{exp}} \right\vert \times 100$
0	0.735	0.735	0
0.1	1.100	0.838	23.8
0.1	1.331	0.838	37.1
0.2	1.032	1.118	8.3
0.3	1.484	1.570	5.8
0.4	–	–	–
0.5	3.229	3.057	5.3
0.5	2.560	3.057	19.4
0.6	5.075	3.883	23.5
0.7	4.740	4.419	6.8
0.8	4.868	4.428	9.0
0.9	3.963	3.944	0.5
1	3.455	3.455	0

The deviations of (η_{exp}) from (η_{theo}) are given in % in the last column. Viscosity data for $x_{Cu} = 0.2$ has been measured by Brillo and co-workers [25]. Experimental data for the pure components are from Refs. [17, 24]

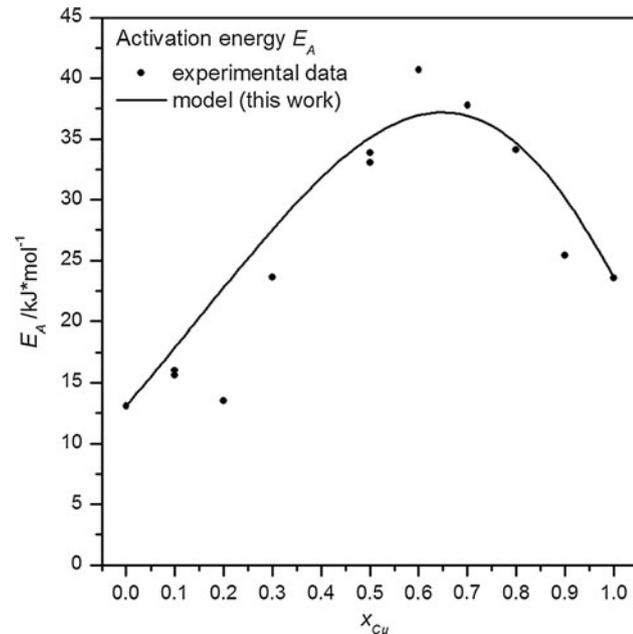activation, and asymptotic viscosity as functions of composition.

Fig. 4 Activation energies, determined from the experimental viscosities in dependence of composition. The *solid curve* represents the results obtained from Eq. (11)
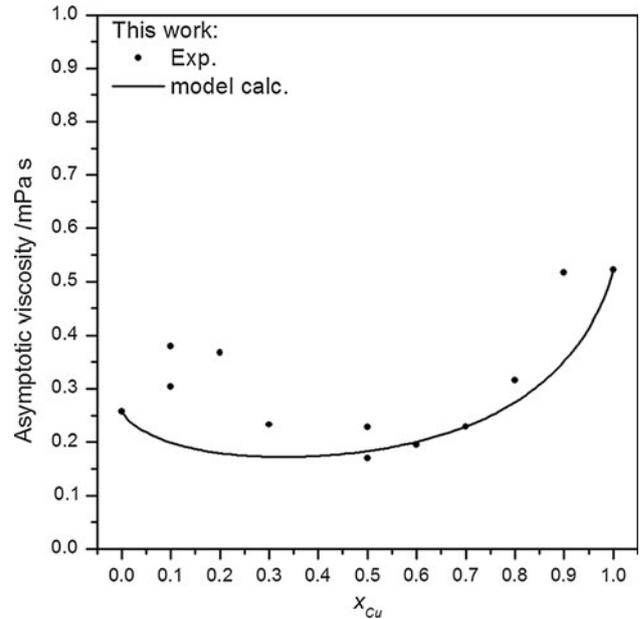

Fig. 5 Asymptotic viscosities η_∞ from viscosity measurements of Al–Cu in comparison to the curve calculated from Eq. (12)

References

1. Brillo J, Brooks R, Egry I, Quested P (2008) High Temp High Press 37:371
2. Egry I (1991) J Mater Sci 26:2997. doi:10.1007/BF01124834
3. Schneider S, Egry I, Seyhan I (2002) Int J Thermophys 23:1241
4. Schmitz J, Brillo J, Egry I, Schmid-Fetzer R (2009) Int J Mater Res 100:1529
5. Brillo J, Plevachuk Yu, Egry I (2010) J Mater Sci 45:5150. doi:10.1007/s10853-010-4512-6
6. Brillo J, Egry I, Westphal J (2008) Int J Mater Res 99:162
7. Brillo J, Egry I (2003) Int J Thermophys 24:1155
8. Jones WRD, Bartlett WL (1952–1953) J Inst Met 81:145
9. Gebhardt E, Becker M, Dorner S (1953) Z Metallkd 44:510
10. Lihl F, Nachtigall E, Schwaiger A (1968) Z Metallkd 59:213
11. Roscoe R (1958) Proc Phys Soc 72:576
12. Friedrichs HA, Ronkow LW, Zhou Y (1997) Steel Res 68:209
13. Plevachuk Y, Sklyarchuk V, Yakymovych A, Eckert S, Willers B, Eigenfeld K (2008) Met Mater Trans A 39:3040
14. Konstantinova NY, Popel PS (2008) J Phys: Conf Ser 98:062022
15. Kehr M, Hoyer W, Egry I (2007) Int J Thermophys 28:1017
16. Dinsdale A, Quested PN (2004) J Mater Sci 39:7221. doi:10.1023/B:JMSC.0000048735.50256.96
17. Brillo J, Brooks R, Egry I, Quested PN (2007) Int J Mater Res 98:457
18. Moelwyn-Hughes EA (1961) Physical chemistry. Pergamon Press, Oxford
19. Kozlov LYa, Romanov LM, Petrov NN (1983) Izv vysch uch zav Chernaya Metallurgiya 3:7
20. Seetharaman S, Sichen D (1994) Met Mater Trans B 25:589
21. Kaptay G (2003) In: Proceedings of microCAD 2003 Conference, Section Metallurgy, University of Miscolc 23
22. Hirai M (1993) ISIJ Int 33:251
23. Ferris D, Quested PN (2002) NPL Report CMMT(A):306
24. Iida T, Guthrie R (1993) The physical properties of liquid metals. Clarendon Press, Oxford

25. Assael MJ, Kakosimos K, Banish RM, Brillo J, Egry I, Brooks R, Quested PN, Mills KC, Nagashima A, Sato Y, Wakeham WA (2006) J Phys Chem Ref Data 52:285

26. Brillo J, Chathoth SM, Koza MM, Meyer A (2008) Appl Phys Lett 93:121905

27. Witusiewicz VT, Hecht U, Fries SG, Rex S (2004) J Alloys Compd 385:133

28. Kehr M, Schick M, Hoyer W, Egry I (2008) High Temp High Press 37:361

29. Walsdorfer H, Arpshofen I, Predel B (1988) Z Metallkd 79:503

30. Brillo J, Bytchkov A, Egry I, Hennet L, Mathiak G, Pozdnyakova I, Price DL, Thiaudiere D, Zanghi D (2006) J Non-Cryst Solids 352:4008

31. Maret M, Pomme T, Pasturel A (1990) Phys Rev B 42:1598

32. Das SK, Horbach J, Koza MM, Mavilla Chatoth S, Meyer A (2005) Appl Phys Lett 86:011918

33. Budai I, Benkö MZ, Kaptay G (2005) Mater Sci Forum 473–474:309

34. Dinsdale AT (1991) CALPHAD 15:317

35. Saunders N (1998) In: Ansara I, Dinsdale AT, Rands MH (eds) COST-507 Thermochemical database for light metal alloys. European Communities, Luxemburg, p 28

36. Redlich O, Kister AT (1948) Ind Eng Chem 40:345

37. Hillert M, Staffansson LI (1970) Acta Chem Scand 24:3618

38. Sundman B, Ågren J (1981) J Phys Chem Solids 42:297