

Calculation of the viscosity of liquid Ag- and Au-based alloys: a pseudopotential approach

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Abstract The viscosity of liquid alloys of Ag–In, Ag–Ge, Ag–Sn, Ag–Sb, and Au–Sn is analyzed in terms of a pseudo-potential method with special emphasis on its variation with the composition. The computational problems associated with the divergence of the key quantities of the theory are solved by presuming the oscillating part of the effective potential to decay exponentially with the distance. The disregard of the influence from neighbor atoms at distances of more than 10 times the hard core diameter is not believed to have an essential impact on the results. All systems under investigation show negative deviations from the additive law which complies well with the experimental findings. The predictions from a semi-empirical model based on simple physical quantities are also taken into consideration. The good overall agreement suggests that the semi-empirical model offers a useful, in many circumstances, more accessible alternative.

Introduction

In the past 10 years, the search of novel lead-free materials for the interconnections in electronic devices has attracted increased attention. A potential group of candidates is based on Sn alloyed with noble metals (Cu, Ag, Au) and other low-melting polyvalent metals (Zn, In, Sb, Bi). Among the physical properties needed to characterize the liquid system, the viscosity is considered as an essential

part of the database. Therefore, in addition to the experimental investigations, the theoretical aspect of the viscosity has become a subject of interest.

The major part of the conventional theories applied to explain the temperature and composition dependence of the viscosity in liquid metals and alloys make use of empirical parameters. One of the probably most successful concepts for alloys is the semi-empirical model proposed by Iida et al. [1, 2], which applies the hard core diameters of the constituent atoms, their masses, and the heat of mixing as input quantities. This three-parameter model turned out to be more successful than the classical Moelwyn–Huges formula [3] which relates the excess viscosity (i.e., the departure from the additive rule) exclusively to the sign of the enthalpy of mixing (H^m) and leads thus to severe contradictions with the experimental findings.

$$\frac{\eta - \eta_{\text{add}}}{\eta_{\text{add}}} = -2 \frac{H^m}{RT} \quad (1)$$

Despite the obvious success of the model of Iida et al. [1, 2], there are also systems (Cu–Pb, Cu–Sb, Cu–Bi, Ag–In, Au–Sn) which are found to be not well represented by the model. Additional improvement could be achieved by adding a term which takes account of the individual volume requirements of the constituents. The advantage of applying such a four-parameter model was discussed previously [4, 5].

As regards the energetics of the systems, it has been emphasized that alloys of noble metals with polyvalent metals have in general a rather ambiguous character. In numerous systems, the enthalpies of mixing are indicative of attractive interactions in noble metal-rich alloys but repulsive in less concentrated alloys. Therefore, it was of interest to use models that do not explicitly involve the enthalpy of mixing as measure of the interactions.

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In a preceding article [5] several binary alloys based on Cu were investigated by the pseudo-potential-based theory of Rice and Alnatt [6], which, in addition to a hard core part, includes a soft attraction due to a weak long-range potential. As matter of fact, the viscosity versus composition curves of Cu–Sb, Cu–Bi, and to some degree also that of Cu–Pb were found to be well reproduced by the theory. Furthermore, it was also observed that the semi-empirical formalism of Iida et al. [4] applied as four-parameter model [5] gave a likewise improved description. In the present article, it will be attempted to extend the considerations to the Ag-based systems Ag–In, Ag–Ge, Ag–Sn, Ag–Sb, and the Au-based system Au–Sn.

Theory

The most crucial ingredient of the theory are the effective potentials. They were constructed in the conventional manner by integration over the normalized wave number characteristics which, in turn, are related to the selected model potentials. Due to their optimum transferability, the pseudo-potentials proposed by Bachelet et al. [7] were assumed to be representative of the systems under investigation. The description concerning the construction of the effective potentials and some other details needed to evaluate the theory are given in [5]. Here, only the most important formulas of the theory will be repeated.

The notations used for the viscosity model in this and the previous paper are the same as that of Kitajima et al. [8] and Bhuiyan et al. [9, 10]. According to the theory of Rice and Alnatt [6], the total viscosity is the sum of a kinetic part η_k , a hard core part $\eta_v(\sigma)$, and a soft part $\eta_v(r > \sigma)$. These are expressed as

$$\eta_k = \sum_{\alpha=1}^2 \sum_{\beta=1}^2 \frac{5k_B T}{8g_{\alpha\beta}(\sigma_{\alpha\beta})} \frac{1 + 4\pi n_{\beta} g_{\alpha\beta}(\sigma_{\alpha\beta}) \sigma_{\alpha\beta}^3 / 15}{(4\pi k_B T / m_{\alpha})^{1/2} \sigma_{\alpha\beta}^2 + [5\zeta_{\alpha}^S / 4m_{\alpha} n_{\beta} g_{\alpha\beta}(\sigma_{\alpha\beta})]} \quad (2)$$

$$\eta_v(\sigma) = \sum_{\alpha=1}^2 \sum_{\beta=1}^2 \left[\frac{5k_B T m_{\alpha\beta} n_{\beta}}{36} \frac{12\pi n_{\alpha} \sigma_{\alpha\beta}^3 / 5g_{\alpha\beta}(\sigma_{\alpha\beta}) + (4\pi n_{\alpha} \sigma_{\alpha\beta}^3 / 5)^2}{(4\pi k_B T / m_{\alpha})^{1/2} \sigma_{\alpha\beta}^2 + [5\zeta_{\beta}^S / 4m_{\alpha} n_{\alpha} g_{\alpha\beta}(\sigma_{\alpha\beta})]} + \frac{8\pi k_B T}{15} \frac{n_{\alpha} n_{\beta} g_{\alpha\beta}(\sigma_{\alpha\beta}) \sigma_{\alpha\beta}^6}{(4\pi k_B T / m_{\alpha})^{1/2} \sigma_{\alpha\beta}^2} \right] \quad (3)$$

$$\eta_v(r > \sigma) = 4\pi \sum_{\alpha=1}^2 \sum_{\beta=1}^2 \frac{m_{\alpha\beta}}{30} \left(\frac{1}{\zeta_{\alpha}^S} + \frac{1}{\zeta_{\beta}^S} \right) X_{\alpha\beta} \quad (\text{for } \alpha \leq \beta) \quad (4)$$

The fundamental input parameters of the theory are the hard sphere diameters ($\sigma_{\alpha\alpha}$, $\sigma_{\beta\beta}$), the atomic masses (m_{α} , m_{β}), the number densities (n_{α} , n_{β}), the respective pair distribution functions $g_{\alpha\beta}(r)$, and the effective pair potentials $u_{\alpha\beta}(r)$. The quantity $m_{\alpha\beta}$ is the reduced mass of m_{α} and m_{β} , $\sigma_{\alpha\beta}$ denotes the arithmetic mean of $\sigma_{\alpha\alpha}$ and $\sigma_{\beta\beta}$. The radial distribution functions $g_{\alpha\beta}(r)$ were approximated by those of a hard sphere mixture [11, 12]. The individual hard core diameters were derived from the empirical relation proposed by Protopapas and Parlee [13].

The friction coefficients ζ_{α}^S and ζ_{β}^S appearing on the right-hand side of Eqs. 2–4 are solved to satisfy the simultaneous equations

$$\zeta_{\alpha}^S = n_{\alpha} \zeta_{\alpha\alpha}^S + n_{\beta} \zeta_{\alpha\beta}^S \quad (5)$$

$$\zeta_{\alpha\beta}^S = \left(\frac{1}{\zeta_{\alpha}^S} + \frac{1}{\zeta_{\beta}^S} \right) c_{\alpha\beta} \quad (6)$$

Computational difficulties arise mainly from the calculation of the quantities $X_{\alpha\beta}$ and $c_{\alpha\beta}$ which affords the integration over the sum of the first derivative $u'_{\alpha\beta}(r)$ and the second derivative $u''_{\alpha\beta}(r)$ of the effective potential with respect to r .

$$X_{\alpha\beta} = n_{\alpha} n_{\beta} \int_{\sigma_{\alpha\beta}}^{\infty} \left[u''_{\alpha\beta}(r) + \frac{4}{r} u'_{\alpha\beta}(r) \right] g_{\alpha\beta}(r) r^4 dr \quad (7)$$

$$c_{\alpha\beta} = \frac{m_{\alpha\beta}}{3} \int_{\sigma_{\alpha\beta}}^{\infty} \left[u''_{\alpha\beta}(r) + \frac{2}{r} u'_{\alpha\beta}(r) \right] g_{\alpha\beta}(r) r^2 dr \quad (8)$$

Due to the weighing factors of r^2 and r^4 , the integrands of $c_{\alpha\beta}$ and $X_{\alpha\beta}$ tend to divergent oscillations as the integration is extended to high values of r . Bhuiyan et al. [9, 10] have considered the effect of truncation at different nodes and found the differences smallest at a cutoff distance of

$r \approx 4.5 \text{ \AA}$. However, a well behaved integration can be obtained if the oscillating part of the effective potential is assumed to decay exponentially from 5 to 10 times the

distance of the hard core diameter. The advantage of this procedure was illustrated in a previous paper [5].

In addition to the full calculation, the experimental results will also be compared with the semi-empirical model of Iida et al. [1, 2]. It is informative to find out whether the model takes advantage of the extension from three to four parameters. The expression for the excess viscosity η^{xs} in relation to the additive viscosity η_{add} formulated as four-parameter model is given as [1, 2, 4]:

$$\frac{\eta^{xs}}{\eta_{add}} = \alpha \frac{x_\alpha x_\beta (\sigma_\alpha - \sigma_\beta)^2}{x_\alpha \sigma_\alpha^2 + x_\beta \sigma_\beta^2} + \beta \left[\left(1 + \frac{x_\alpha x_\beta (m_\alpha^{1/2} - m_\beta^{1/2})^2}{(x_\alpha m_\alpha^{1/2} + x_\beta m_\beta^{1/2})^2} \right)^{1/2} - 1 \right] + \gamma \frac{2H^m}{RT} + \delta \frac{2x_\alpha x_\beta |V_\alpha - V_\beta|}{V_\alpha + V_\beta} \quad (9)$$

Here, σ_α and σ_β were identified with the ionic diameters. V_α and V_β stand for the atomic volumes of the elements, x_α and x_β are their atomic fractions, and T denotes the temperature. The enthalpies of mixing H^m needed to evaluate the third term of Eq. 9 were taken from the literature [14]. In general, these data did not refer to the reference temperature of the viscosity; however, as a first estimate they were assumed to be temperature independent. As a matter of fact, some of them have a considerable temperature dependence, but in view of the scatter of the experimental data and the small value of the weighing factor γ of the H^m -term in Eq. 9, this error was found to be very small.

Based on a statistical consideration of the existing viscosity data, the best values of the weighing factors were found to be [4]:

$$\alpha = -4.05, \beta = 12.0, \gamma = -0.115, \delta = -2.67.$$

The values quoted in the original three-parameter model [1, 2] are different since the last term in Eq. 9 had not been considered ($\alpha = -5.0, \beta = 2.0, \gamma = -0.12, \delta = 0.0$). Both sets of parameters will be used in order to work out the differences.

Results and discussion

As regards the energetics of the system, the Ag-based systems considered here display the typical feature of alloys between noble and polyvalent metals. In a very characteristic manner, the enthalpy of mixing follows an S-shaped curve which is well pronounced in Ag–In, Ag–Ge, and Ag–Sn but less indicated in Ag–Sb (Fig. 1). The exothermic part on the Ag-rich side points to attractive interactions (i.e., tendency of compound formation),

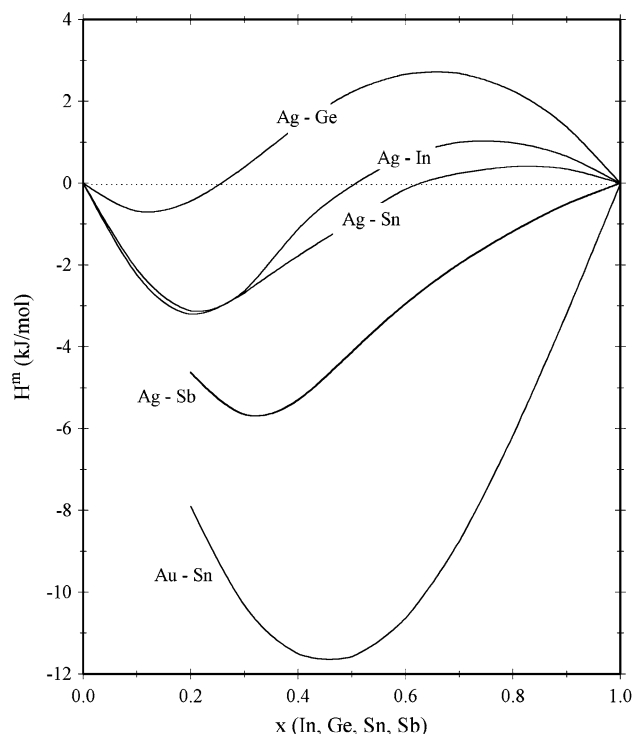


Fig. 1 Enthalpies of mixing in various noble metal-based liquid alloys taken from Ref. [14]: Ag–In (1100 K), Ag–Ge (1250 K), Ag–Sn (1250 K), Ag–Sb (1250 K), and Au–Sn (823 K)

whereas the endothermic part on the polyvalent metal side implies repulsive interactions (i.e., tendency of demixing). Au–Sn is one among the Au-based systems that does not show this typical behavior.

It was clear from the beginning that the excess viscosity of the present systems is not at all correlated with the enthalpy of mixing as expected at first sight from Moelwyn–Huges’ formula [3]. According to Eq. 1, exothermic systems ($H^m < 0$) are expected to exhibit increased viscosities (i.e., positive deviations from linearity) while endothermic systems ($H^m > 0$) should have viscosities smaller than the additive value. The systems considered here are predominantly exothermic; however, the measurements revealed negative excess viscosities in all systems under investigation. Furthermore, the S-formed shape of the enthalpy curve characteristic of the systems Ag–In, Ag–Ge, and Ag–Sn is apparently not reflected by the experimental viscosities (Figs. 2–6).

In the model of Iida et al. [1, 2], the enthalpy of mixing is given less weight, consequently the complexity of the enthalpy of mixing curve is not expected to be directly reflected in the viscosity isotherms. In Ag–In, Ag–Sn, and Au–Sn, the curves calculated by the four-parameter model are closer to the experimental viscosities than the three-parameter model, while in Ag–Ge and Ag–Sb there are no obvious improvements. Here, it has to be emphasized that

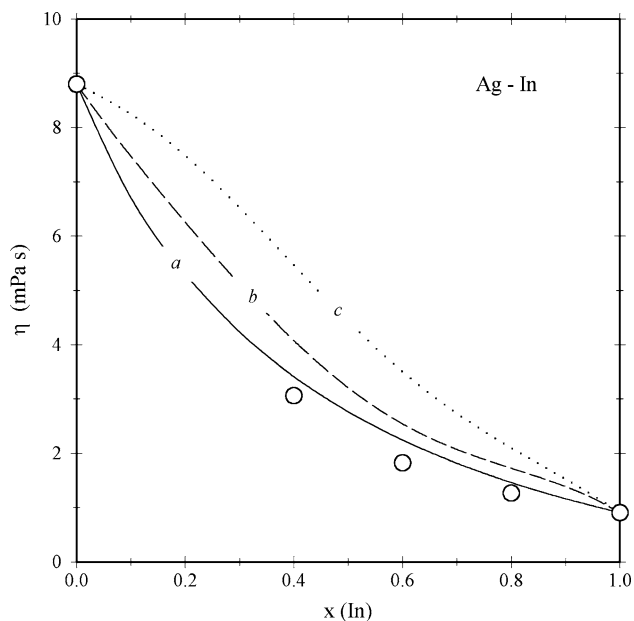


Fig. 2 Composition dependence of the viscosity in liquid Ag–In alloys at 900 K: *a*, full calculation; *b*, four-parameter model; *c*, three-parameter model; O, experimental

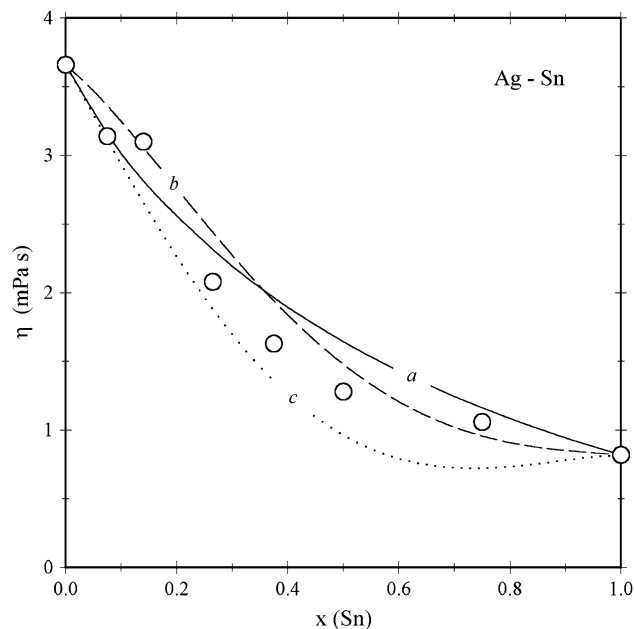


Fig. 4 Composition dependence of the viscosity in liquid Ag–Sn alloys at 1273 K: *a*, full calculation; *b*, four-parameter model; *c*, three-parameter model; O, experimental

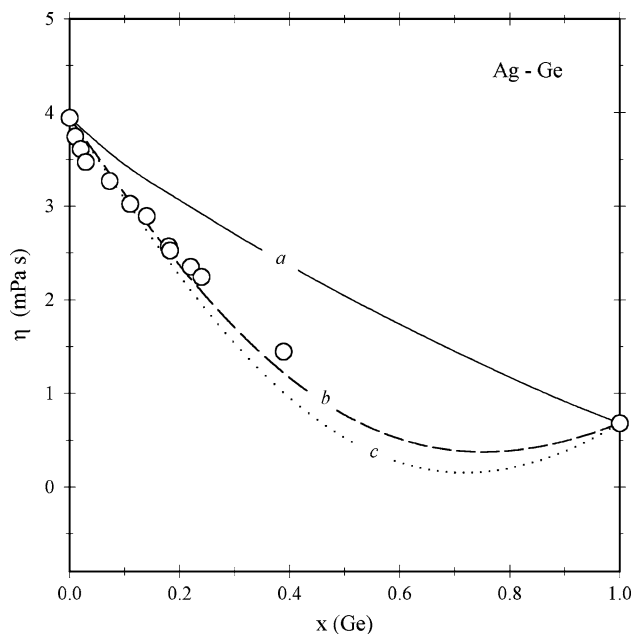


Fig. 3 Composition dependence of the viscosity in liquid Ag–Ge alloys at 1260 K: *a*, full calculation; *b*, four-parameter model; *c*, three-parameter model; O, experimental

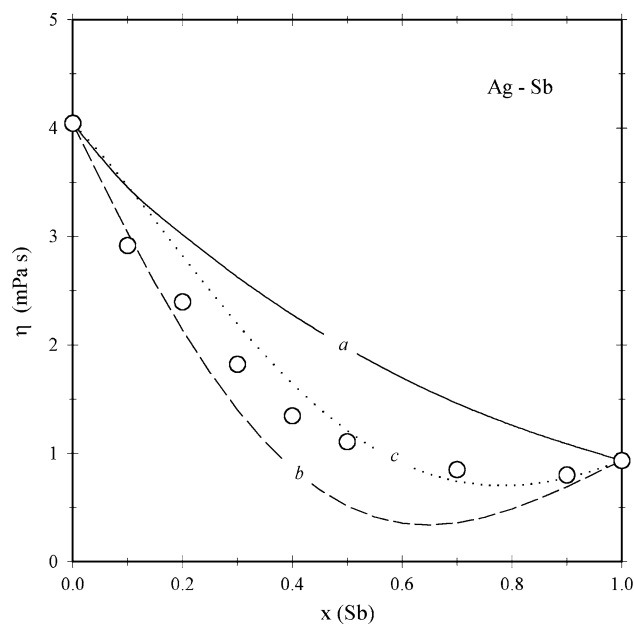


Fig. 5 Composition dependence of the viscosity in liquid Ag–Sb alloys at 1273 K: *a*, full calculation; *b*, four-parameter model; *c*, three-parameter model; O, experimental

the extent of deviations from the additive law shown for Ag–In and Au–Sn depend also on the value extrapolated for liquid Ag and liquid Au to a reference temperature far below their melting points (1234 K for Ag and 1336 K for Au). As a matter of fact, the extrapolated viscosities have to be considered as crude estimates based on the assumption of an Arrhenius-like temperature dependence.

The focus of the semi-empirical model [1, 2, 4] is on the excess viscosity and not on the absolute value. The full calculation based on the pseudo-potential approach, on the other hand, should be capable to give absolute values, provided sufficient attention is given to appropriate input quantities. The most decisive simplifications are presumably the choice of model potential, the application of hard

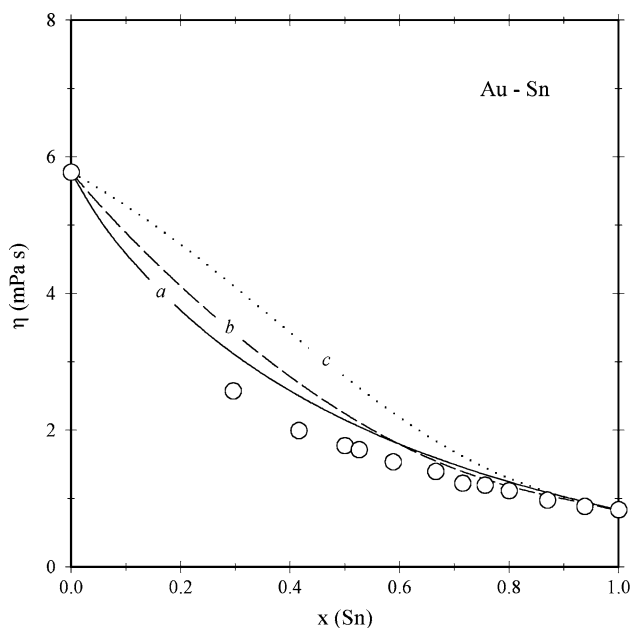


Fig. 6 Composition dependence of the viscosity in liquid Au–Sn alloys at 1173 K: *a*, full calculation; *b*, four-parameter model; *c*, three-parameter model; O, experimental

sphere distribution functions, the integration limit $\sigma_{\alpha\beta}$, and to some extent also the arbitrariness in the modification of the long-range part of the effective potential that was introduced in order to achieve a controlled integration. In view of the circumstances it is not surprising to find the calculated viscosities different from the experimental values.

A summary of the calculations obtained for the pure elements is given in Table 1. The total viscosities of Ag and Au are found to be considerably smaller than the experimental ones, while those of the polyvalent elements turned out to be larger than actually observed.

Table 1 Individual contributions to the viscosity of some liquid metals obtained from the pseudopotential approach

	<i>T</i> (K)	Viscosity (mPa s)				Exp.	$f_{\alpha\alpha}, f_{\beta\beta}$
		η_k	$\eta_v(\sigma)$	$\eta_v(r > \sigma)$	η_{tot}		
Ag	900	0.092	1.756	0.579	2.427	(8.80)	–
	1273	0.110	1.441	0.520	2.071	3.66	1.767
Au	1173	0.141	2.255	0.766	3.162	5.78	1.828
In	900	0.038	0.491	0.673	1.202	0.91	0.703
Ge	1273	0.057	1.044	0.482	1.583	0.85	0.524
Sn	1173	0.044	0.491	0.653	1.188	0.84	0.524
	1273	0.044	0.462	0.681	1.187	0.82	0.528
Sb	1273	0.100	0.771	0.156	1.028	0.93	0.457

Note: *T*, reference temperature; η_k , kinetic contribution; $\eta_v(\sigma)$, hard core part; $\eta_v(r > \sigma)$, soft part; η_{tot} , total viscosity; Exp., experimental values; $f_{\alpha\alpha}, f_{\beta\beta}$, adjustment factors

The main concern of the present investigation was on the deviations from the additive law; therefore, the calculated viscosities were adjusted so as to match with the experimental values of the elemental components. Kitajima et al. [8] have adjusted their calculations to the experimental viscosities by fitting their potential to the viscosities at the two limits of the concentration range. In the present investigation, the calculated viscosities were corrected with the factor $(f_{\alpha\alpha}, f_{\beta\beta})^{1/2}$, which implies that all terms in Eqs. 2–4 originating from the pure elements were corrected with their individual factors ($f_{\alpha\alpha}$ or $f_{\beta\beta}$), whereas the terms due to the mixture ($\alpha \neq \beta$) were corrected with their geometric mean. The factors introduced for the adjustment of the pure elements ($f_{\alpha\alpha}, f_{\beta\beta}$) are listed in the last column of Table 1. They are found to be large for the noble metals ($f_{\alpha\alpha} \approx 2.0$), but smaller than unity for the alloying elements ($f_{\beta\beta} \approx 0.5\text{--}0.7$).

The total viscosities of the noble metals are found to be dominated by a large hard core part $\eta_v(\sigma)$ while in some of the polyvalent metals (In, Sn) the soft part $\eta_v(r > \sigma)$ gives definitely the greater contribution. The calculations of Bhuiyan et al. [9, 10] for alloys of Ag–In and Ag–Sn yielded hard core contributions much higher than observed in the present calculation. Some of the results are listed in Table 2. It is interesting to note that even in In- or Sn-rich alloys their hard core part (1.65 mPa s for 80 at% In, and 1.05 mPa s for 90 at% Sn) exceeds the soft part (0.257 and 0.336 mPa s, respectively) by a factor of 3–6, whereas the present calculations yielded comparable amounts of around 0.55–0.75 mPa s. Kitajima [15], on the other hand, found the hard core contribution both in In and Sn

Table 2 Viscosities of some representative liquid alloys of Ag–In and Ag–Sn calculated by different authors on the basis of the pseudopotential approach

Alloy	Viscosity (mPa s)				<i>T</i> (K)
	η_k	$\eta_v(\sigma)$	$\eta_v(r > \sigma)$	η_{tot}	
Ag–In					
30 at% In ^a	0.086	1.06	0.744	1.90	900
30 at% In ^b	0.140	2.45	0.291	2.88	973
80 at% In ^a	0.053	0.588	0.739	1.38	900
80 at% In ^b	0.097	1.65	0.257	2.00	623
100 at% In ^d	0.051	0.210	1.477	1.74	433
Ag–Sn					
20 at% Sn ^a	0.129	1.50	0.863	2.50	1173
20 at% Sn ^c	0.135	2.12	0.675	2.91	1023
90 at% Sn ^a	0.055	0.549	0.695	1.30	1173
90 at% Sn ^c	0.067	1.05	0.336	1.45	572
100 at% Sn ^d	0.019	0.215	1.059	1.290	523

^a This work

^{b,c,d} Literature data taken from Refs. [9, 10, 15]

(≈ 0.21 mPa s) to be even remarkably smaller than the soft part (1.48 mPa s for In and 1.06 mPa s for Sn) (Table 2).

These discrepancies underline that the outcome of the theory is highly sensitive to the choice of input quantities. To some extent the disagreement is due to different reference temperatures (cf. Table 2); however, the most decisive quantity of the theory is presumably the choice of model potential. Kitajima [15] applied empty-core potentials [16] adapted so as to match the experimental viscosities of the elements while Bhuiyan et al. [9, 10] favored a Bretonnet–Silbert type potential [17, 18]. In the present investigation, due to the argument of transferability, the more elaborate pseudopotentials put forward by Bachelet et al. [7] were considered to be most suitable.

Ag–In

The measurements of Iida [19, 20] were performed far below the melting point of Ag and on alloys with less than 60 at% Ag. In Fig. 2, the experimental points are combined with the value of 8.80 mPa s estimated for undercooled liquid Ag according to the data compiled in the literature [21]. It is the question whether this extremely high value has physical significance. The highest experimental values reported for liquid Ag are 3.67 mPa s at 1273 K [22] or 4.07 mPa s at 1250 K [23], both far away from the extrapolated value. The experimental viscosity isotherm in Fig. 2 has therefore a highly speculative character, but the principal shape might be considered realistic. The experimental curve, as far as known, is well reproduced by the full calculation (curve a). The four-parameter model (curve b) yields a likewise good description of the viscosity, much better than the original three-parameter model (curve c).

Ag–Ge

The experimental investigation of Martin-Garin et al. [23] covers only a part of the system from Ag up to 40 at% Ge and are therefore not very reliable as far as the complete viscosity isotherm is considered (Fig. 3). The full calculation (curve a) suggests negative excess viscosities much smaller than indicated by the experimental points. It is interesting that the semi-empirical models (curves b and c) yield good agreement with a minimum of the viscosity predicted around 70 at% Ge. Strictly, the minimum is due to the extremely small ionic diameter of Ge^{4+} (1.06 Å) compared to that of Ag^+ (2.52 Å), which gives the first term in Eq. 9 a dominating role. It is unclear whether the minimum has physical significance or is simply due to an overestimation of this term in the model. In this

exceptional case, an extension of the measurements toward the Ge-rich side would make the situation clear.

Ag–Sn

The experimental points given in Fig. 4 are those reported by Gebhardt et al. [24]. The shape of their viscosity isotherm is in good accordance with the calculations. The isotherm deduced from the four-parameter model (curve b) is practically the same as that of the full calculation (curve a) and even the three-parameter model (curve c) gives an acceptable description of the system.

Ag–Sb

In this system, the agreement with the experiment is only of qualitative character. The excess viscosities calculated with the full theory (curve a) are less negative than those observed by Nakajima [25], whereas the four-parameter model (curve b) overestimates the deviations from additivity. In this particular case, the three-parameter model (curve c) gives the best accordance with the experiment.

Au–Sn

Figure 6 shows the experimental results observed by Gebhardt et al. [26]. The extrapolation of the viscosity of pure liquid Au to a temperature 160 K below the melting point yields a value of 5.78 mPa s [21], which is a reasonable value if the experimental viscosity 5.13 mPa s measured at 1373 K [22] is taken into consideration. Despite the lack of experimental data from 70–100 at% Au, the deviations are apparently strongly negative. This is supported by the calculations. Both the full calculation (curve a) as well as the four-parameter model (curve b) are in reasonable accordance with the experimental findings; only the three-parameter model (curve c) fails to represent the system. Additional measurements aimed at closing the gap of experimental results on the Au-rich side are highly recommended.

Conclusion

The particular pseudo-potential method used in this and the preceding paper is capable of explaining the experimental viscosity isotherms in various Cu-, Ag-, and Au-based binary alloys. The basic assumption is the exponential decay of the oscillating part of the effective potential at distances of more than 15–20 Å. The approach

underestimates the influence of neighboring atoms from larger distances but guarantees also the convergence of the integrations needed to evaluate the theory. The improvements are primarily concerned with the divergent character of the quantities $c_{\alpha\beta}$ and $X_{\alpha\beta}$.

Apparently, the semi-empirical theory applied as four-parameter model gives also a good explanation of the experimental excess viscosities. In view of the computational difficulties associated with the pseudo-potential approach and the necessity to bring the calculated values into scale with the experimental viscosities of the elements, the application of the semi-empirical model appears to be a good first, easily accessible alternative. All in all, it seems that the application in terms of a four-parameter model is more adequate than its use in the original three-parameter version.

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