

COMPARISON OF DIFFERENT THEORETICAL MODELS TO EXPERIMENTAL DATA ON VISCOSITY OF BINARY LIQUID ALLOYS

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Abstract. Six different theoretical equations are compared in the present paper with experimental data, measured for 28 binary liquid metallic systems. General conclusions are drawn on the ability of the different theoretical models to describe the concentration and temperature dependence of the viscosity of liquid alloys. A new equation is derived, being able to predict the viscosity in multi-components alloy even if the viscosities of the pure components are not known.

Introduction

Viscosity of binary and multi-component liquid alloys is one of their basic physical properties, being also important for the design of materials technologies. Six different models have been published so far to describe the concentration dependence of the viscosity of liquid alloys through their thermodynamic properties [1-7]. In recent years, one, or another model was chosen (with no explanation of its choice) to estimate the viscosity of some metallic systems [8-15]. However, the six existing models were compared to experimental data systematically only in a few cases: for the Ag-Sb [16], Ag-Cu [17], Ag-In and In-Sb [18] systems. The aim of this paper is to collect reliable experimental data for a large number of systems and to compare them with the existing models. The systems will be grouped and analyzed according to their phase diagrams [19].

Only those literature models are considered here, which can be applied without knowing the actual value of the viscosity at any composition of the binary alloy. The models are divided into two groups. First, all models will be considered, which use the known viscosity values of the pure components as initial parameters. Second, the models without this initial information will be discussed. A new model will be developed for this latter case.

Models with known viscosities of pure components, as initial parameters

The Moelwyn-Hughes (MH) equation [1] was the first to take into account that the viscous flow becomes more difficult when the cohesion energy of the alloy is increased:

$$\eta = (x_1 \cdot \eta_1 + x_2 \cdot \eta_2) \cdot \left(1 - 2 \cdot x_1 \cdot x_2 \cdot \frac{\Omega}{R \cdot T} \right) \quad (1)$$

where η and η_i - dynamic viscosity of the alloy and of pure phase i ($i = 1, 2$), (Pas), x_i - mole fraction of component i , $R = 8.315$ J/molK, the universal gas constant, T - absolute temperature (K), Ω - the mixing enthalpy parameter of the alloy ($\Delta H = x_1 x_2 \Omega$), (J/mol).

The Iida-Ueda-Morita (IUM) equation [2, 3] in addition to excess thermodynamic terms takes into account the influence of the differences in atomic mass (m_i , kg/atom) and size (d_i , the double of Pauling's ionic radius of the ions, m) of the components:

$$\eta = (x_1 \cdot \eta_1 + x_2 \cdot \eta_2) \cdot \left\{ 2 \cdot \left[1 + \frac{x_1 \cdot x_2 \cdot (\sqrt{m_1} - \sqrt{m_2})^2}{(x_1 \cdot \sqrt{m_1} + x_2 \cdot \sqrt{m_2})^2} \right]^{1/2} - 1 - \frac{5 \cdot x_1 \cdot x_2 \cdot (d_1 - d_2)^2}{x_1 \cdot d_1^2 + x_2 \cdot d_2^2} - \Delta \right\} \quad (2)$$

Δ - is the thermodynamic correction, defined in two different ways [2, 3]:

$$\Delta = 0.12 \cdot \frac{\Delta H}{R \cdot T} \quad \text{or} \quad \Delta = 0.12 \cdot \frac{\Delta G^E}{R \cdot T} \quad (2.a-b)$$

where $\Delta G^E = \Delta H - T\Delta S^E$, i.e. the integral excess Gibbs energy, expressed through the heat of mixing (ΔH) and excess entropy of mixing (ΔS^E).

The Kozlov-Romanov-Petrov (KRP) equation [4] is derived in a theoretical way, and finds the correlation in semi-logarithmic coordinates:

$$\ln \eta = \sum_{i=1}^n x_i \cdot \ln \eta_i - \frac{\Delta H}{3 \cdot R \cdot T} \quad (3)$$

The Seetharaman-Du Sichen (SDS) equation [6] is based on the Eyring equation, with describing the activation energy of the alloy by the semi-empirical Eq.(4.a):

$$\eta = h \cdot N_{Av} \cdot \frac{\rho}{M} \cdot \exp\left(\frac{\Delta G^*}{R \cdot T}\right) \quad (4)$$

where h – is the Planck constant ($6.626 \cdot 10^{-34}$ J s), N_{Av} – is the Avogadro number ($6.022 \cdot 10^{23}$ 1/mol), ΔG^* - the Gibbs energy of activation of the viscous flow (J/mol) defined as [6]:

$$\Delta G^* = \sum_{i=1}^2 x_i \cdot \Delta G_i^* + R \cdot T \cdot \sum_{i=1}^2 x_i \cdot \ln x_i + 3 \cdot R \cdot T \cdot x_1 \cdot x_2 + \Delta H - T \cdot \Delta S^E \quad (4.a)$$

ΔG_i^* – the Gibbs energy of activation of the viscous flow in pure component i , calculated from the measured viscosity of the pure component at the given temperature (see Eq.(4)):

$$\Delta G_i^* = R \cdot T \cdot \ln\left(\frac{\eta_i \cdot M_i}{h \cdot N_{Av} \cdot \rho_i}\right) \quad (4.b)$$

The Kaptay (K) equation [7] is a modification of Eq.(4), taking into account the theoretical relationship between the cohesion energy of the alloy and the activation energy of viscous flow, leading to the following value of parameter $\alpha = 0.155 \pm 0.015$:

$$\eta = \frac{h \cdot N_{Av}}{\sum_i x_i \cdot V_i + \Delta V^E} \cdot \exp\left[\frac{\sum_i x_i \cdot \Delta G_i^* - \alpha \cdot \Delta H}{R \cdot T}\right] \quad (5)$$

where ΔV^E – the excess molar volume upon alloy formation (m^3/mol), which can be neglected for simplicity, when experimental data are not available.

Models with un-known viscosities of pure components

This group of equations provides a possibility to calculate the viscosity of alloys even in the case, when the viscosity of one, or more components are not known in their pure state at the temperature of interest. It is clear, however, that this strength of such models is also their weakness, as in some cases even the viscosity of the pure metal is predicted with a relatively high uncertainty.

The Hirai (H) equation [5] is a semi-empirical extrapolation of Andrade's equation, suggested originally for pure liquid metals:

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

where ρ - density of the alloy (kg/m^3), M – average atomic weight of the alloy (kg/mol), T_m – melting point, i.e. the liquidus temperature of the alloy (K). Thus, in order to apply the Hirai equation, the liquidus surface (temperature) should be known as function of composition, what is not always the case for multi-component alloys. It should also be mentioned that the Hirai equation leads to unreasonable discontinuities on the viscosity versus composition curve at eutectic and monotectic compositions. Moreover, it cannot be used above the miscibility gaps in monotectic systems, as the liquidus temperature is not known.

The combination of the unified equation with any of the equations (1-5). The following unified equation has been elaborated recently [40], to describe the viscosity of all pure liquid metals as a function of temperature with an uncertainty of $\pm 20\%$:

$$\eta_i = A \cdot \frac{M_i^{1/2}}{V_i^{2/3}} \cdot T^{1/2} \cdot \exp\left(B \cdot \frac{T_{m,i}}{T}\right) \quad (7)$$

with η_i (Pas), M_i (kg/mol), V_i (m^3/mol), $T_{m,i}$ (K) being the dynamic viscosity, atomic mass, molar volume and melting point of the given metal i . The above equation was tested on 101 measured points of 15 selected liquid metals, and the average values of the generally valid parameters were found as: $A = (1.80 \pm 0.39) \cdot 10^{-8} (\text{J/Kmol}^{1/3})^{1/2}$, $B = 2.34 \pm 0.20$. Parameter B in Eq.(7) has been theoretically defined as [40]:

$$B \equiv q \cdot \frac{\Delta Z}{Z} \quad (7.a)$$

where q is a semi-empirical parameter ($q \approx 25.4 \pm 2$ [41]), ΔZ is the number of broken bonds during viscous flow ($\Delta Z \approx 1$), Z is the average coordination number in liquid metals ($Z \approx 11$). The physical sense of parameter q is related to the cohesion energy in pure liquid metals, defined as [41]:

$$\Delta_c U_i = -q \cdot R \cdot T_{m,i} \quad (7.b)$$

If the viscosity of one or more pure components of a multi-component system are not known at the temperature of interest, Eq.(7) can be used to estimate the un-known values, and the estimated values can be used in the combination with any of the Eq-s (1-5) to calculate the concentration dependence.

A new equation has been derived by us (called later as Budai-Benkő-Kaptay (BBK) model), being a binary- and multi-component extension of Eq.(7). The molar mass, the molar

volume and the cohesion energy of pure liquid metals i in Eq.(7) were extended to multi-component alloys by the following obvious relationships:

$$M = \sum_i x_i \cdot M_i \quad (8.a)$$

$$V = \sum_i x_i \cdot V_i + \Delta V^E \quad (8.b)$$

$$\Delta_c U_i = -q \cdot R \cdot \sum_i x_i \cdot T_{m,i} + \Delta H \quad (8.c)$$

where the excess molar volume (ΔV^E) and the integral heat of mixing (ΔH) are concentration and temperature dependent quantities, to be found from independent experiments or theories. Substituting Eq-s (8.a-c) into Eq.(7), the following unified equation for the viscosity of multi-component liquid alloys can be found:

$$\eta_i = A \cdot \frac{\left(\sum_i x_i \cdot M_i \right)^{1/2}}{\left(\sum_i x_i \cdot V_i + \Delta V^E \right)^{2/3}} \cdot T^{1/2} \cdot \exp \left[\frac{B}{T} \cdot \left(\sum_i x_i \cdot T_{m,i} - \frac{\Delta H}{q \cdot R} \right) \right] \quad (8.d)$$

Description of the experimental data and the method of comparison

Altogether experimental data for 28 binary liquid alloy systems have been found in the literature, which describe the concentration dependence of a given liquid alloy in the whole concentration region. The phase diagrams and thermodynamic properties of the alloys were taken from [19-20]. The viscosities for pure metals were taken from the same original papers on the viscosity of the binary alloys. The molar volume (or density) of pure liquid metals were taken from [25]. The excess molar volumes of all systems were taken as zero, for simplicity. The systems are grouped according to their phase diagram types in Table 1. In Table 1 the maximum deviation between experimental and theoretical values are indicated in relative %. At the end of each sub-table, the average deviation, the average of the absolute deviations, and the ranking of different methods (based on the average of the absolute deviations) are given. The average absolute deviations are summarized in Table 2. The final ranking is found using the weight factor, according to the number of the systems of the same type.

Table 1. Maximum deviation of model calculations from experimental data (%) at given temperature. Abbreviations: MH = Moelwyn-Hughes model [1], IUM = Iida-Ueda-Morita model [2,3], KRP = Kozlov-Romanov-Petrov model [4], SDS = Seetharaman-Du Sichen model [6], K = Kaptay model [7], H = Hirai model [5], BBK = Budai-Benkő-Kaptay model (this paper)

Table 1.a. Systems with solid solutions

System	T [K]	MH	IUM	KRP	SDS	K	H	BBK
Ag-Au [21]	1373	+80	-5	-5	-10	+1	-5	+12
Au-Cu [2]	1473	+67	+2	0	-37	0	-24	+22
Bi-Sb [2]	973	-10	+2	+2	+15	+2	+68	+87
Co-Fe [33]	1873	+29	-6	-5	+3	+5	-26	-10
Average		42	-2	-2	-7	2	3	28
Abs. average		47	4	3	16	2	31	33
Ranking		7	3	2	4	1	5	6

Table 1.b. Eutectic systems

System	T [K]	MH	IUM	KRP	SDS	K	H	BBK
Ag-Cu [22]	1373	-75	+5	+5	+10	+2	-32	+14
Ag-Cu [22]	1473	-70	-5	0	+7	+2	-33	+16
Ag-Cu [22]	1573	-67	-6	-2	+5	-2	-32	+13
Al-Zn [27]	973	-50	+30	+25	+35	+26	-30	+55
Bi-Cu [28]	1416	-72	+76	+18	+23	+9	+95	+61
Bi-Sn [25]	673	-15	-11	-13	-10	-13	+34	+50
Cd-Pb [32]	+623	-100	+28	+23	+35	+21	-12	+30
Pb-Sb [38]	973	+9	-1	+7	+13	+7	+73	+94
Pb-Sn [38]	673	-47	-7	-12	-11	-11	-30	-10
Pb-Sn [38]	773	-39	-1	-9	-6	-7	-28	-7
Pb-Sn [38]	873	-35	-4	-10	-8	-8	-25	-6
Sn-Zn [32]	723	-100	+70	51	+67	+44	-35	+50
Average		-55	15	7	13	5	-5	30
Abs. average		57	20	15	19	13	38	34
Ranking		7	4	2	3	1	6	5

Table 1.c. Monotectic systems

System	T [K]	MH	IUM	KRP	SDS	K	H*	BBK
Bi-Zn [31]	873	-140	+60	+40	+51	+34	+120	+40
Bi-Zn [31]	923	-130	+48	+38	+50	+30	+112	+37
Average		-135	54	39	51	32	116	39
Abs. average		135	54	39	51	32	116	39
Ranking		7	4	2	3	1	6	5

*the temperature along the miscibility gap was taken into account, as the liquidus T is unknown

Table 1.d. Systems with congruently melting compound(s)

System	T [K]	MH	IUM	KRP	SDS	K	H	BBK
Al-Cu [4]	1473	+90	-46	-45	-50	-46	-47	-29
Bi-In [29]	573	+72	+5	+9	-9	+5	+40	+53
Bi-In [29]	623	+72	+5	+10	-9	+5	+36	+50
Bi-In [29]	673	+68	+6	+11	-9	+6	+33	+45
Bi-In [29]	723	+65	+7	+12	-9	+7	+30	+42
Bi-Tl [30]	623	+165	+9	+18	+7	+14	+35	+54
Bi-Tl [30]	673	+165	+8	+18	+7	+15	+35	+51
Bi-Tl [30]	723	+158	+9	+20	+9	+16	+30	+51
Cu-Sb [25]	1373	+185	+78	+45	+35	+31	+50	+95
Hg-Na [2]	643	+200	-38	-9	-69	-47	-19	-55
Mg-Pb [37]	973	+230	+15	+30	-15	+17	+20	+56
Mg-Pb [37]	1073	+205	+12	+29	-20	+13	+20	+56
Mg-Pb [37]	1173	+180	+14	+24	-26	-15	+20	+54
Average		143	6	13	-12	2	22	40
Abs. average		143	19	22	21	18	32	53
Ranking		7	2	4	3	1	5	6

Table 1.e. Systems with compound(s), dissociating by a peritectic reaction

System	T [K]	MH	IUM	KRP	SDS	K	H	BBK
Ag-In [23]	1250	+190	+31	+14	+8	+12	-20	+4
Ag-Sb [24]	1273	+100	+30	+50	+65	+62	+68	+110
Ag-Sb [25]	1273	+150	+60	+70	+76	+67	+65	+110
Ag-Sn [26]	1273	+130	+40	+40	+45	+40	-18	+43
Ag-Sn [26]	1373	+110	+30	+36	+40	+32	-20	+37
Ag-Sn [26]	1473	+100	-23	+32	+35	+27	-22	+34
Cd-Sn [32]	623	-53	+41	+40	+45	+38	+9	+46
Cd-Sn [39]	623	-60	+29	+23	+36	+22	-16	+19
Cu-Sn [34]	1373	+86	+18	-14	-25	-19	-41	-22
Cu-Sn [34]	1473	+76	+30	-11	-22	-16	-40	-20
Cu-Sn [34]	1573	+74	+28	+10	-20	-15	-40	-21
In-Pb [35]	623	-31	-4	-4	+5	-2	-9	+10
In-Pb [35]	673	-31	+5	-2	+5	-2	-10	+7
In-Pb [35]	723	-29	+1	-5	+2	-4	-10	+6
In-Pb [35]	773	-28	+1	-7	-4	-7	-13	+5
In-Sn [29]	523	+38	+28	+28	+30	+29	+20	+47
In-Sn [29]	573	+35	+26	+26	+28	+26	+15	+38
In-Sn [29]	623	+33	+20	+25	+27	+25	+14	+37
In-Sn [29]	673	+32	+23	+25	+29	+25	+15	+33
In-Sn [29]	723	+31	+22	+25	+28	+24	+15	-31
K-Na [36]	376,3	-39	-18	-8	+6	0	-37	-9
K-Na [36]	394,5	-37	-20	-10	+7	0	-34	-8
K-Na [36]	420,1	-34	-25	-11	+6	-1	-31	-5
K-Na [36]	440,4	-32	-28	-12	+5	0	-29	-5
K-Na [36]	466	-30	-31	-13	+5	0	-27	-5
Sn-Tl [30]	673	-30	-7	-13	-8	-12	-30	-12
Sn-Tl [30]	623	-32	-7	-10	-7	-10	-29	-12
Sn-Tl [30]	723	-28	-9	-10	-8	-10	-29	-10
Average		25	10	11	16	12	-10	15
Abs. average		60	23	21	22	19	26	27
Ranking		7	4	2	3	1	5	6

Table 2. Average absolute deviation of different models for predicting the viscosity of binary liquid alloys of different phase diagram types (abbreviations see above Fig-s 1)

System type	n	MH	IUM	KRP	SDS	K	H	BBK
Solid solution	4	47	4	3	16	2	31	33
Eutectic	12	57	20	15	19	13	38	14
Monotectic	2	135	54	39	51	32	116	38
Peritectic compound	28	60	23	21	22	19	26	27
Congruent compound	13	143	19	22	21	18	32	53
Weighed average		79	21	19	22	17	33	31
Ranking		7	3	2	4	1	6	5

Conclusions

- i. Among the models with known viscosities of pure components as initial parameters, the MH model provides unreasonable predictions in most of the cases, so it should not be used. The models SDS, IUM, KRP and K provide similar results. As the closest agreement between experiments and predictions for all types of phase diagrams is reached using the K-model, this model should be preferred when an unknown system is modeled (see Eq-s.(5, 4b)).
- ii. Among the models with un-known viscosities of pure components as initial parameters, the H and BBK models provide similar results. However, the H-model provides unreasonable discontinuities at eutectic compositions, and cannot be used in principle for monotectic systems. That is why, the BBK model is suggested by us to be used when the viscosities of none of the pure components are known. Alternatively, Eq.(7) is suggested to be used to predict the unknown viscosities of pure components. These predicted values should be combined with known viscosities of other pure components, and finally the K-model (Eq-s (5, 4.b)) should be used to obtain the concentration dependence of viscosity of binary and multi-component alloys.
- iii. The performance of the most preferred models is the best for systems with relatively simple phase diagrams, showing a relatively small deviation from ideality (solid solution type and eutectic type), while the agreement becomes worse when the system deviates more from ideality both into positive (monotectic systems) and negative (systems with compounds) directions.
- iv. The disagreement in certain cases becomes unacceptable for systems with congruently melting compound(s) in the phase diagram. This indicates the existence of associates (complexes, molecules) in the liquid alloy, which change all physical properties of the alloy in a hardly predictable manner. For systems with congruently melting compounds none of the existing models can be recommended for reliable predictions. Instead, the following procedure is suggested. First, the viscosity of the "pure" associate can be estimated using Eq.(7), if the molar volume and the congruent melting point of the associate is known. Then, the associate model [42-46] should be used to find the mole fractions of un-associated atoms and that of the associates. These model mole fractions and the viscosity of the associate from Eq.(7) should be combined with the viscosities of pure components into Eq.(5). This algorithm is expected to provide a better estimation for systems with stable associates, usually showing a large positive deviation from the linear composition dependence of viscosity (see especially the Al-Cu and Hg-Na systems in Table 1.d).
- v. The next generation of models to predict the viscosity of multi-component liquid alloys as function of temperature and concentration should take into account: i). the heat of mixing of the alloy (see Eq.5), ii). the inner segregation of the components into the sliding planes of the alloy (see [16]), iii). the excess molar volume of the alloy (see Eq.6 and the free volume theory), iv). and the formation of associates in the liquid alloy.

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