Reference Correlation for the Density and Viscosity of Eutectic Liquid Alloys Al+Si, Pb+Bi, and Pb+Sn

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In this paper, the available experimental data for the density and viscosity of eutectic liquid alloys Al+Si, Pb+Bi, and Pb+Sn have been critically examined with the intention of establishing a reference standard representation of both density and viscosity. All experimental data have been categorized as primary or secondary according to the quality of measurement, the technique employed, and the presentation of the data, as specified by a series of carefully defined criteria. The proposed standard reference correlations for the density of liquid Al+Si, Pb+Bi, and Pb+Sn are, respectively, characterized by deviations of 2.0%, 2.9%, and 0.5% at the 95% confidence level. The standard reference correlations for the viscosity of liquid Al+Si, Pb+Bi, and Pb+Sn are, respectively, characterized by deviations of 7.7%, 14.2%, and 12.4% at the 95% confidence level. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4750035]

Key words: bismuth; density; eutectic; lead; metal; reference correlations; tin; viscosity.

CONTENTS

List of Tables

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List of Figures

1. Introduction

Following the need for reference values of the density and viscosity of liquid metals identified over several years, a project was initiated by the International Association for Transport Properties, IATP (former Subcommittee on Transport Properties of the International Union of Pure and Applied Chemistry, IUPAC) in 2006 to evaluate critically the density and the viscosity of selected liquid metals. Thus,

- in 2006, reference values for the density and viscosity of liquid aluminum and iron were published,[1] as a result of a project supported by IUPAC.
- Following this, in 2010, values for the density and viscosity for liquid copper and tin were proposed.[2] That work had also been supported by IUPAC.
- In 2011, the work was continued and reference correlations of the density and viscosity of liquid bismuth, nickel, lead, silver and antimony were proposed,[3] while in 2012 the work was concluded with liquid cadmium, cobalt, gallium, indium, mercury, silicon, thallium, and zinc.[4]

For the remaining liquid metals in the periodic table, very limited information is available in the literature.

The present work proposes reference correlations for the liquid eutectic alloys Al+Si, Pb+Bi, and Pb+Sn. These three eutectic alloys were selected because measurements for their density and viscosity are available from several sources. The following should also be noted:

- Alloy Al+Si shows a eutectic concentration at 12.0% by mass (11.53% by atom) of Si, and it is employed by the metal casting industry and in functionally graded materials (FGM).
- Alloy Pb+Bi shows a eutectic concentration at 55.5% by mass (56.25% by atom) of Bi, and is employed as a coolant in primary circuits in nuclear reactors.
- Alloy Pb+Sn shows a eutectic concentration at 61.9% by mass (73.9% by atom) of Sn, and is employed in the electronic industry.

2. Primary and Secondary Data

According to the recommendation adopted by the Subcommittee of Transport Properties (now known as The International Association for Transport Properties) of the International Union of Pure and Applied Chemistry, experimental data can be placed into two categories according to the quality of the data: primary and secondary data. As already discussed,[1–4] the primary data are identified by the following criteria:[5]

(i) Measurements must have been made with a primary experimental apparatus, i.e., one for which a complete working equation is available.
(ii) The form of the working equation should be such that sensitivity of the property measured to the principal variables does not magnify the random errors of measurement.
(iii) All principal variables should be measurable to a high degree of precision.
(iv) The published work should include some description of purification methods and a guarantee of the purity of the sample.
(v) The data reported must be unsmoothed data. While graphs and fitted equations are useful summaries for the reader, they are not sufficient for standardization purposes.
(vi) The lack of accepted values of the density and viscosity of standard reference materials implies that only absolute, and not relative, measurement results can be considered.
(vii) Explicit quantitative estimates of the uncertainty of reported values should be given, taking into account the precision of experimental measurements and possible systematic errors.
(viii) Owing to the desire to produce reference values of low uncertainty, limits must be imposed on the uncertainty of the primary data sets. These limits are determined after critical evaluation of the existing data sets.

These criteria have been successfully employed to propose standard reference values for the viscosity and thermal conductivity of fluids over a wide range of conditions, with uncertainties in the region of 1%.

However, in the case of the liquid metals and their alloys, it was argued that these criteria needed to be relaxed slightly, since the uncertainty of the measurements is generally much higher, primarily owing to (i) the difficulties associated with the techniques employed at such high temperatures and (ii) the purity of the liquid metal sample which can be strongly affected by the surrounding atmosphere and the container used for the melt.

3. Density

3.1. Experimental techniques

Among the experimental work identified for the density of molten materials, a large number of techniques have been employed to measure the density of eutectic liquid alloys Al+Si, Pb+Bi, and Pb+Sn. Methods employed include: Archimedean; pycnometric; bubble-pressure; sessile-drop; large-drop; levitation; and gamma radiation attenuation. These methods have been presented in our previous compilations[1–4] and will not be further discussed here; nothing significantly different has been applied in the work reviewed here.

It should also be noted that, although some investigators have noticed a hysteresis in the density values between heating and cooling, recent work[6] has shown that this effect disappears upon proper mixing.

3.2. Data compilation

Table 1 presents the data sets found for the measurement of the density of eutectic liquid alloys Al+Si, Pb+Bi, and Pb+Sn. In this table, the purity of the components, the composition of the alloy, the technique employed and the uncertainty quoted are also presented. Furthermore, the form in which the data are presented and the temperature range covered are also noted. The data sets have been classified into primary and secondary sets according to the criteria presented in Sec. 2 and in conjunction with a review of the techniques described in our previous work.[1–4] More specifically, the following can be noted.

- Al±Si: Six investigators reported density measurements for this eutectic liquid alloy. The measurements of Magnusson and Arnberg[7] and Siddiqui *et al.*[8] were performed by the Archimedean technique with low uncertainty and were considered as primary data. The measurements of Wang *et al.*[9] obtained in a sessile-drop instrument with low uncertainty were also considered as primary data together with the electromagnetic levitation measurements of Schmitz *et al.*[10] and the γ-ray measurements of Popel *et al.*[11] Finally, the measurements of Peijie *et al.*[12] performed in a γ-ray instrument were considered as secondary data, because they were shown in a very small graph, as the authors were only interested in the investigation of the effect on the density of adding Ce in this alloy.
- Pb±Bi: Density measurements have been reported by five investigators. The measurements of Stankus *et al.*[13] and Yagodin *et al.*[14] were performed in an absolute way, in γ-ray instruments, with low uncertainty and were thus considered as primary data. The measurements of Alchagirov *et al.*[15] were obtained in an absolute pycnometer with very low uncertainty and were also part of the primary data. As primary data, the sessile-drop measurements of Kazakova *et al.*[16] were also included as they covered a wide range, even if their uncertainty was worse. Finally, the measurements of Plevachuk *et al.*,[17] performed in a large-drop instrument, were considered as secondary data, as the authors themselves recognize that their data are lower than all other data.
- Pb±Sn: The low-uncertainty measurements of Khairulin and Stankus,[6] performed in a γ-ray instrument, of Wang and Xian,[18] performed in an Archimedean apparatus, and of Thresh and Crawley,[19] performed in a pycnometer, were all considered as primary data. The measurements of Gebhardt and Kostlin,[20] obtained in an Archimedean apparatus, and of Fischer and Phillips,[21] obtained in a maximum bubble-pressure instrument, were also part of the primary data set. The measurements of Gasior *et al.*[22] obtained in a dilatometer were considered as secondary data, as they show a distinctively different slope than the all other measurements. The γ-ray measurements of Popel *et al.*[23] were also considered in this case as secondary, as they deviated from all other data sets, systematically and in excess of the quoted uncertainty.

3.3. Density reference correlation

The primary density data for the liquid eutectic alloys, shown in Table 1, were employed in a linear regression analysis to represent the density at 0.1 MPa as a function of the temperature. Since the quoted uncertainties of all works were of similar magnitude, the data were weighted only according to the number of points. The following equations were obtained for the density, ρ (kg m^{-3}), as a function of the absolute temperature, T (K),

$$\rho = c_1 - c_2 T, \tag{1}$$

and the coefficients c_1 (kg m^{-3}) and c_2 (kg m^{-3} K^{-1}) are shown for each liquid eutectic alloy in Table 2. In the same table, the percentage deviation (2σ) of each equation at the 95% confidence level is also shown.

Figures 1–3 show the primary data and their percentage deviations from the above equation for each of the three liquid eutectic alloys. The dashed vertical line shows the melting point for each alloy. The following can be observed:

- In the case of Al+Si (Fig. 1), although most investigators quote uncertainty below 1%, their measurements differ among themselves by up to 1.5%.
- In the case of Pb+Bi, the measurements of Kazakova *et al.*[16] and Yagodin *et al.*[14] are further apart than the other two sets. However, as already stated, there is no justification not to consider them as primary data. Therefore, the deviations are within 2.9% at the 2σ confidence level. It should be pointed out that in the case of the Pb-Bi eutectic,

TABLE 1. Data sets considered for the density of liquid eutectic Al+Si, Pb+Bi, and Pb+Sn.

First author	Publ. year	Technique employed[a]	Purity[b] (mass %)	Composition[c] (mass %)	Uncertainty quoted (%)	No. of data	Form of data[d]	Temperature range (K)
				Al+Si				
Primary data								
Schmitz[10]	2012	EML (Abs)	99.999 99.999	12.0	1.0	20	P	951–1601
Magnusson[7]	2001	Archimedean (Abs)	99.999 99.99	11.6	0.3	8	P	871–1073
Wang[9]	2001	Sessile drop (Abs)	99.995 99.99	12.5	0.5	17	P	928–1454
Popel[11]	1987	γ-ray	99.999 99.999	12.7	0.07	17	D	872–1725
Siddiqui[8]	1987	Archimedean (Abs)	99.74 na	11.0	na	5	P	858–965
Secondary data								
Peijie[12]	1996	γ-ray (Abs)	na na	11.7	0.1	4	D	973–1273
				Pb+Bi				
Primary data								
Stankus[13]	2006	γ-ray (Abs)	99.998 99.98	55.5	0.3–0.4	115	D	404–1224
Yagodin[14]	2005	γ-ray (Abs)	na na	55.4	0.5	12	E	400–950
Alchagirov[15]	2003	Pycnometer (Abs)	na na	55.5	0.1	83	P	410–726
Kazakova[16]	1984	Sessile drop	99.999 99.999	56.7	2.0	9	E	400–1200
Secondary data								
Plevachuk[17]	2011	Large drop	na na	56.0	1.5	7	E	400–700
				Pb+Sn				
Primary data								
Khairulin[6]	2007	γ-ray (Abs)	99.99 99.99	61.9	0.2	118	D	453–1036
Wang[18]	2005	Archimedean (Abs)	99.99 99.99	60.0	0.4	5	P	463–570
Thresh[19]	1970	Pycnometer (Abs)	99.997 99.999	62.5	0.05	8	E	463–820
Gebhardt[20]	1957	Archimedean (Abs)	99.99 99.9	61.9	na	9	P	523–973
Fischer[21]	1954	Maximum bubble pressure (Abs)	99.998 99.98	62.05	na	12	D	400–950
Secondary data								
Popel[23]	1985	γ-ray (Abs)	99.9 99.95	61.9	0.07	25	D	420–950
Gasior[22]	2001	Dilatometer (Abs)	99.995	61.9	0.5	15	P	564–1200

[a]Abs = absolute; EML = electromagnetic levitation; Rel = relative.
[b]Purity refers to 1st and 2nd component, respectively.
[c]Composition refers to mass percentage of second component.
[d]D = diagram; E = equation; P = points.

a correlation was also proposed by Sobolev[24] in 2010. This correlation is in excellent agreement with the present one.

- In the case of Pb+Sn, the deviations were less than 0.4% at the 2σ confidence level.

Finally, in Table 3, density values calculated with the use of Eq. (1) are shown.

TABLE 2. Temperature range, coefficients, and deviations at the 95% confidence level of Eq. (1).

	T_{range} (K)	c_1 (kg m^{-3})	c_2 (kg m^{-3} K^{-1})	Deviation (2σ) (%)
Al+Si	858–1700	2603	0.241	2.0
Pb+Bi	400–1225	10922	1.096	2.9
Pb+Sn	400–1040	8472	0.810	0.4

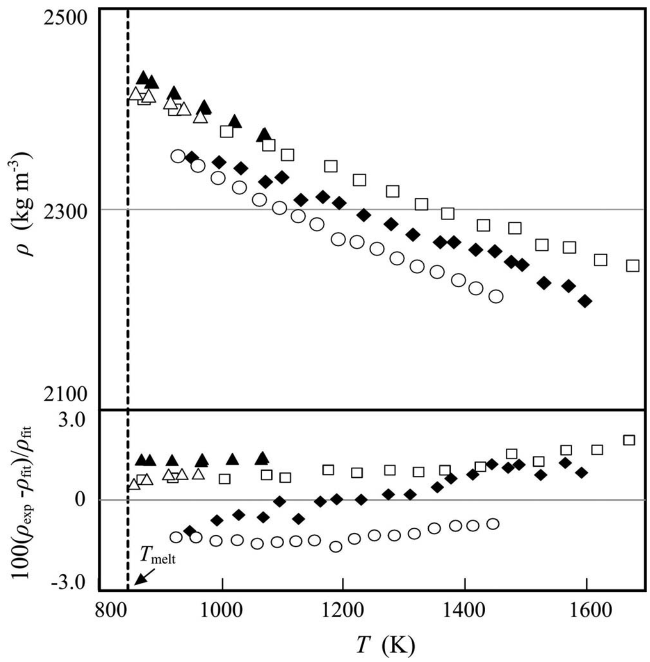

FIG. 1. Primary density data and their percentage deviations from Eq. (1) for eutectic liquid alloy Al+Si as a function of temperature. Schmitz *et al.*[10] (♦), Magnusson and Arnberg[7] (▲), Wang *et al.*[9] (○), Popel *et al.*[11] (□), and Siddiqui *et al.*[8] (Δ).

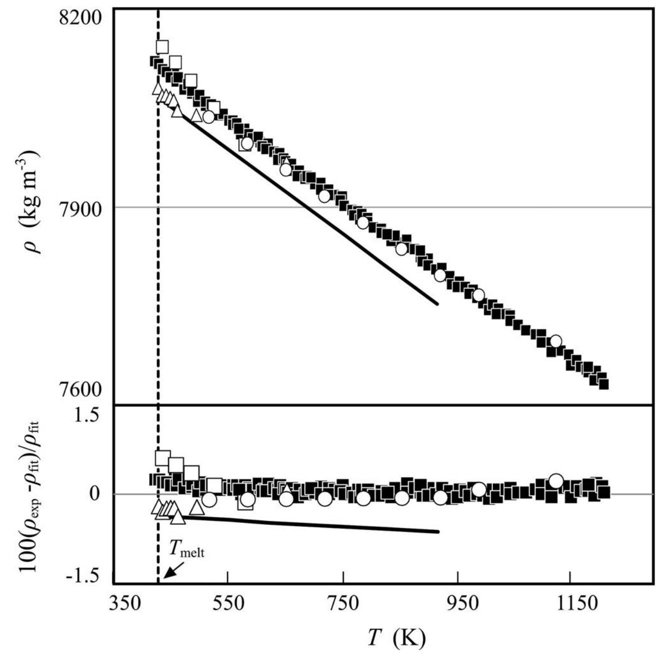

FIG. 3. Primary density data and their percentage deviations from Eq. (1) for eutectic liquid alloy Pb+Sn as a function of temperature. Khairulin and Stankus[6] (■), Wang and Xian[18] (□), Thresh and Crawley[19] (—), Fischer and Phillips[21] (Δ), and Gebhardt and Kostlin[20] (○).

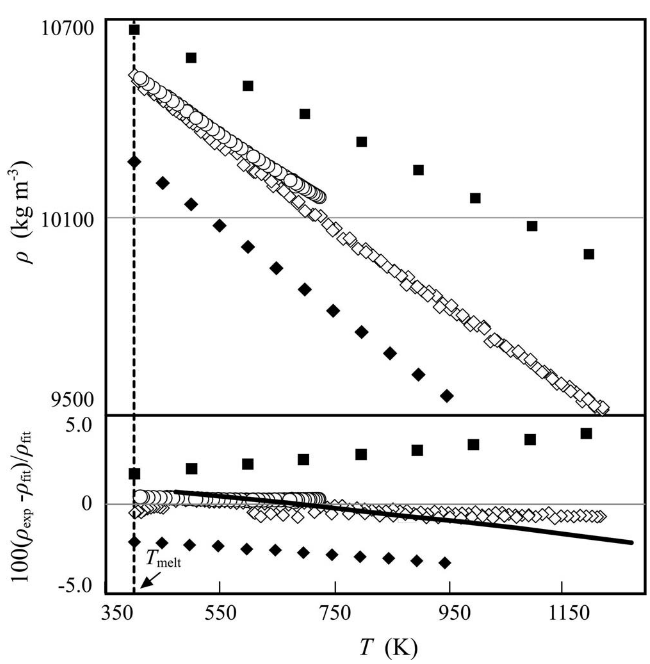

FIG. 2. Primary density data and their percentage deviations from Eq. (1) for eutectic liquid alloy Pb+Bi as a function of temperature. Stankus *et al.*[13] (◊), Yagodin *et al.*[14] (■), Alchagirov *et al.*[15] (○), and Kazakova *et al.*[16] (♦).

4. Viscosity

4.1. Experimental techniques

There exist a large number of methods to measure the viscosity of liquids, but those suitable for liquid metals are limited by the low viscosities of metals (of the order of 1–10 mPa s), their chemical reactivity, and generally high melting points. In the case of the three eutectic alloys examined, three techniques in total were employed: the oscillating cup, γ-rays, and the Archimedean technique. These methods have been presented in our previous compilations[1–4] and will not be discussed further here.

4.2. Data compilation

Table 4 presents the data sets found for the measurement of the viscosity of eutectic liquid alloys Al+Si, Pb+Bi, and Pb+Sn. In the table, for every data set, the technique employed, the purity of the components, the composition of the alloy, the uncertainty quoted, the form of the data presented, the number of data points as well as the temperature range to which they refer, are also shown. The data sets have been classified into primary and secondary sets according to the criteria presented in Sec. 2 and in conjunction with the techniques described previously.[1–4]

TABLE 3. Recommended values for the density and viscosity of liquid eutectic alloys Al+Si, Pb+Bi, Pb+Sn.

T (K)	ρ (kg m^{-3})	η (mPa s)	T (K)	ρ (kg m^{-3})	η (mPa s)	T (K)	ρ (kg m^{-3})	η (mPa s)
	Al+Si			Pb+Bi			Pb+Sn	
850	2398	0.919	400	10484	3.549	400	8148	2.986
900	2386	0.850	500	10374	2.380	500	8067	2.162
1000	2362	0.718	600	10264	1.824	600	7986	1.743
1100	2338	0.626	700	10155	1.508	700	7905	1.494
1200	2314	0.558	800	10045	1.307	800	7824	1.331
1300	2290	0.506	900	9936	1.170	900	7743	1.217
1400	2266		1000	9826	1.071	1000	7662	1.133
1500	2242		1100	9716	0.996	1100	7581	
1600	2217		1200	9607	0.937			
1700	2193		1300	9497				

TABLE 4. Data sets considered for the viscosity of liquid eutectic Al+Si, Pb+Bi, and Pb+Sn.

First author	Publ. year	Technique employed[a]	Purity[b] (mass %)	Composition[c] (mass %)	Uncertainty quoted (%)	No. of data	Form of data[d]	Temperature range (K)
				Al+Si				
Primary data								
Song[25]	2009	Oscillating cup (Abs)	99.999 99.999	12.0	na	9	D	872–1273
Moraru[26]	2007	Oscillating cup (Abs)	99.7 99.7	12.0	na	14	D	862–982
Geng[27]	2005	Oscillating cup (Abs)	99.7 99.9	12.5	5.0	15	D	928–1454
Secondary data								
Peijie[12]	1996	Oscillating cup (Abs)	na na	11.7	0.1	4	D	973–1273
				Pb+Bi				
Primary data								
Gusachev[28]	2011	Oscillating cup (Abs)	na na	55.5	2.0	16	P	350–1100
Plevachuk[29]	2008	Oscillating cup (Abs)	na na	56.0	3.0	153	D	400–996
Kaban[30]	2004	Oscillating cup (Abs)	99.999 99.999	55.9	5.0	136	G	407–1072
Kaplun[31]	1979	Oscillating cup (Abs)	99.991 99.992	55.5	5.0	98	P	394–1181
Nikol’skii[32]	1959	Oscillating cup (Abs)	na na	56.5	na	14	P	423–1073
Secondary data								
Wu[33]	2007	Oscillating cup (Abs)	99.95 99.98	55.2	1.0	14	D	398–806
				Pb+Sn				
Primary data								
Sklyarchuk[34]	2011	γ-ray (Abs)	99.99 99.999	61.9	5.0	34	D	464–797
Plevachuk[35]	2005	Oscillating cup (Abs)	99.999 99.999	61.9	3.0	5	D	453–750
Thresh[19]	1970	Oscillating cup (Abs)	na na	61.9	0.5	2	D	623, 823
Kanda[36]	1968	Oscillating cup (Abs)	99.97 99.999	61.9	1.0	7	D	494–770
Toye[37]	1958	Oscillating cup (Abs)	99.97 99.998	61.9	0.5	3	D	456–700
Gebhardt[20]	1957	Oscillating cup (Abs)	99.99 99.99	62.05	4.0	10	P	473–973

TABLE 4. Data sets considered for the viscosity of liquid eutectic Al+Si, Pb+Bi, and Pb+Sn.—Continued

First author	Publ. year	Technique employed[a]	Purity[b] (mass %)	Composition[c] (mass %)	Uncertainty quoted (%)	No. of data	Form of data[d]	Temperature range (K)
Jones[38]	1957	Archimedean (Abs)	99.99 99.9	61.9	na	9	P	523–973
Fischer[21]	1954	Oscillating cup (Abs)	99.998 99.98	62.05	na	19	D	458–664
Secondary data								
Wu[33]	2007	Oscillating cup (Abs)	99.95 99.98	61.9	1.0	24	D	456–894
Yao[39]	1952	Oscillating cup (Abs)	99.9885 99.9962	61.8	na	15	D	460–726

[a]Abs = absolute; Rel = relative.
[b]Purity refers to 1st and 2nd component, respectively.
[c]Composition refers to mass percentage of second component.
[d]D = diagram; E = equation; P = points.

In the case of the viscosity data sets and in relation to the discussion of Sec. 4.1, the following points can be noted:

– Al±Si: Four investigators reported viscosity measurements for this eutectic liquid alloy. The measurements of Song *et al.*,[25] Moraru,[26] and Geng *et al.*[27] were all performed in oscillating-cup instruments and composed the primary data set. The measurements of Peijie *et al.*,[12] performed also in an oscillating-cup instrument, were considered as secondary data, because they were shown only in a very small graph, and the authors were only interested in an investigation of the effect on viscosity when adding Ce to this alloy.
– Pb+Bi: In the case of the measurement of the viscosity of eutectic Pb+Bi, all investigators employed the oscillating-cup technique. From the six investigators that reported viscosity measurements, Gusachev *et al.*,[28] Plevachuk *et al.*,[29] Kaban *et al.*,[30] Kaplun *et al.*,[31] and Nikol'ski *et al.*[32] were all considered as primary data. Gusachev *et al.*[28] and Nikol'ski *et al.*[32] reported kinematic viscosities, and thus the density equation proposed in Sec. 3 was used to convert them to dynamic viscosities. The measurements of Wu *et al.*[33] were not included in the primary data set, as they were far higher than the measurements of all other investigators.
– Pb+Sn: 10 investigators reported measurements of the viscosity of this eutectic alloy. Eight of them were included in the primary data sets. The measurements of Plevachuk *et al.*,[35] Thresh and Crawley,[19] Kanda and Colburn,[36] Toye and Jones,[37] Gebhardt and Kostlin,[20] and Fisher and Phillips[21] were all performed in absolute oscillating-cup instruments and were part of the primary data sets. The measurements of Slyarchuk *et al.*,[34] performed in a γ-ray instrument, and the measurements of Jones and Davies,[38] performed by the Archimedean technique, also formed part of the primary data sets. Among the sets of data to be considered secondary, we include the measurements of Wu *et al.*[33] and Yao and Kondic,[39] whose results were much higher than all other investigators; in the case of Wu *et al.*,[33] this was also the case in Pb+Bi measurements above, while in the case of Yao and Kondic[39] the same trend was also noticed in previous evaluations.[3,4]

4.3. Viscosity reference correlation

The primary viscosity data for eutectic liquid alloys Al+Si, Pb+Bi, and Pb+Sn, shown in Table 4, were employed in a regression analysis as a function of the temperature. The data were weighted according to the number of points. The following equations were obtained for the viscosity, η (mPa s), as a function of the absolute temperature, T (K),

$$\log_{10}(\eta/\eta^\circ) = -a_1 + \frac{a_2}{T}, \tag{2}$$

where $\eta^\circ = 1$ mPa s, and the coefficients a_1 (−), and a_2 (K) are shown for each liquid alloy in Table 5. In the same table, the percentage deviation (2σ) of each equation at the 95% confidence level is also shown.

Figures 4–6 show the primary viscosity data and their percentage deviations from Eq. (2) for each liquid alloy. The dashed vertical line shows the melting point for each alloy. The following can be observed for these three figures:

- In almost all cases, the differences between authors are much larger than the claimed uncertainties, so that the overall uncertainty of the correlation is higher.
- In the case of Al+Si eutectic alloy, more viscosity measurements are required.

TABLE 5. Temperature range, coefficients, and deviations at the 95% confidence level of Eq. (2).

	T_{range} (K)	a_1 (−)	a_2 (K)	Deviation (2σ) (%)
Al+Si	860–1275	0.8022	658.34	7.7
Pb+Bi	350–1185	0.3173	346.95	14.2
Pb+Sn	450–975	0.2266	280.69	12.4

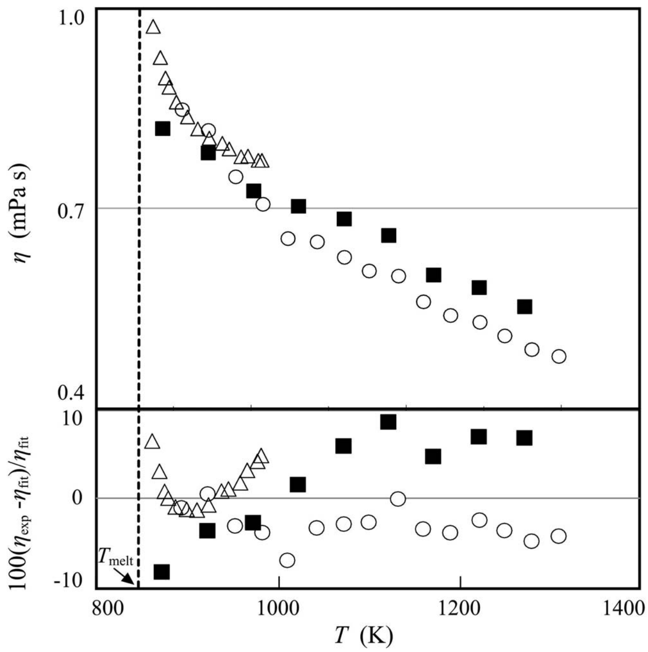

FIG. 4. Primary viscosity data and their percentage deviations from Eq. (2) for eutectic liquid alloy Al+Si as a function of temperature. Song *et al.*[25] (■), Moraru[26] (Δ), and Geng *et al.*[27] (○).

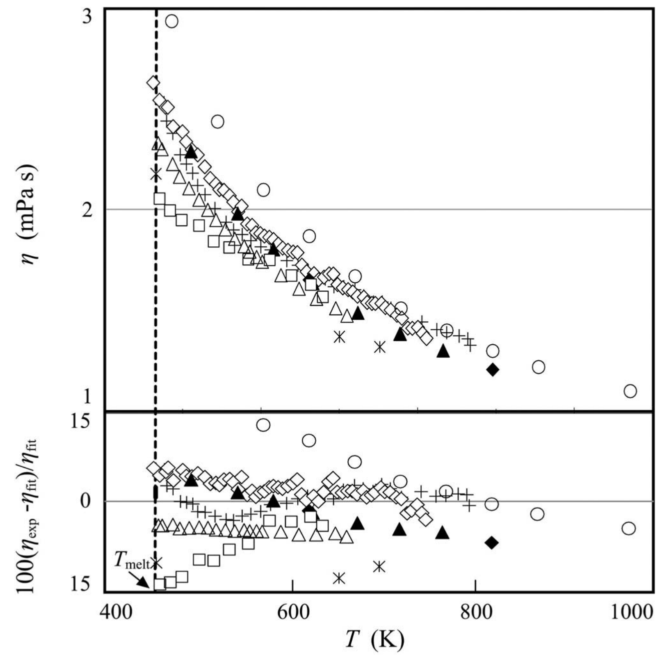

FIG. 6. Primary viscosity data and their percentage deviations from Eq. (2) for eutectic liquid alloy Pb+Sn as a function of temperature. Slyarchuk *et al.*[34] (+), Plevachuk *et al.*[35] (◊), Thresh and Crawley[19] (♦), Kanda and Colburn[36] (▲), Toye and Jones[37] (⁎), Gebhardt and Kostlin[20] (○), Jones and Davies[38] (□), and Fisher and Phillips[21] (Δ).

- In the case of Pb+Bi and Pb+Sn eutectic alloys, better measurements of lower uncertainty are required.

Viscosity values calculated from the above equation are contained in Table 3.

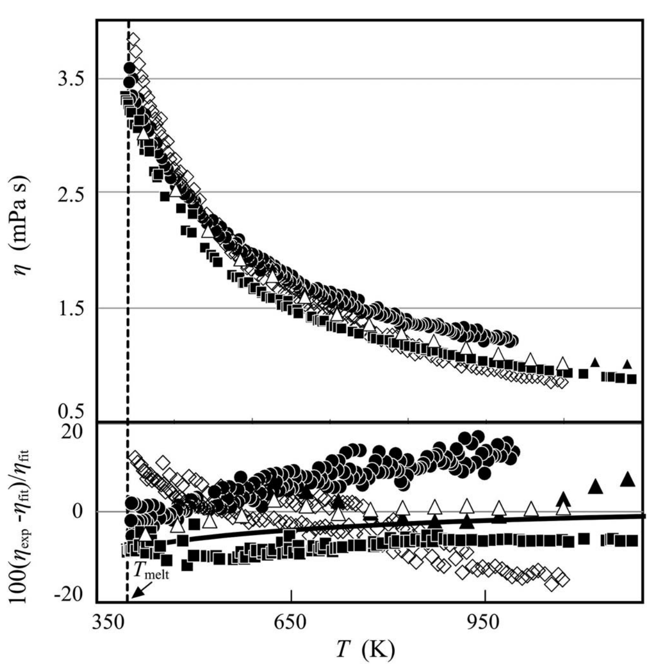

FIG. 5. Primary viscosity data and their percentage deviations from Eq. (2) for eutectic liquid alloy Pb+Bi as a function of temperature. Gusachev *et al.*[28] (▲), Sobolev[24] (—), Plevachuk *et al.*[29] (●), Kaban *et al.*[30] (◊), Kaplun *et al.*[31] (■), and Nikol'ski *et al.*[32] (Δ).

5. Conclusions

The available experimental data for the density and viscosity of eutectic liquid alloys Al+Si, Pb+Bi, and Pb+Sn have been critically examined with the intention of establishing a density and a viscosity standard. All experimental data have been categorized into primary and secondary data according to the quality of measurement, the technique employed, and the presentation of the data, as specified by a series of criteria. The proposed standard reference correlations for the density of eutectic liquid alloys Al+Si, Pb+Bi, and Pb+Sn are characterized by deviations of 2.0%, 2.9%, and 0.5% at the 95% confidence level. The standard reference correlations for the viscosity of liquid Al+Si, Pb+Bi, and Pb+Sn are characterized by deviations of 7.7%, 14.2%, and 12.4% at the 95% confidence level, respectively.

It is obvious that much more work, and certainly measurements with lower uncertainty, needs to be carried out in this area. The reference values proposed by this work represent the best that can be done with the present literature. Nevertheless, the deviations of the proposed equations are quite high and high enough, we judge, to be of concern in practical applications.

Finally, we note that the proposed correlations are for vapor-liquid saturation conditions. Although in some applications, such as the flow in a tube or a nozzle, the pressure is higher than the saturation pressure, the pressure dependences of the density and the viscosity of liquid metal alloys are not sufficiently high that the variation exceeds the uncertainty in the correlations reported here.

Acknowledgments

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6. References

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