Reference Data for the Density and Viscosity of Liquid Cadmium, Cobalt, Gallium, Indium, Mercury, Silicon, Thallium, and Zinc

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The available experimental data for the density and viscosity of liquid cadmium, cobalt, gallium, indium, mercury, silicon, thallium, and zinc have been critically examined with the intention of establishing both 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 liquid cadmium, cobalt, gallium, indium, silicon, thallium, and zinc are characterized by percent deviations at the 95% confidence level of 0.6, 2.1, 0.4, 0.5, 2.2, 0.9, and 0.7, respectively. In the case of mercury, since density reference values already exist, no further work was carried out. The standard reference correlations for the viscosity of liquid cadmium, cobalt, gallium, indium, mercury, silicon, thallium, and zinc are characterized by percent deviations at the 95% confidence level of 9.4, 14.0, 13.5, 2.1, 7.3, 15.7, 5.1, and 9.3, respectively.
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Key words: cadmium; cobalt; density; gallium; indium; melt; mercury; reference data; silicon; tin; thallium; viscosity; zinc

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1. Introduction

There is a continual increase in the use of mathematical models to simulate a variety of processes involving liquid metals such as shape-casting; primary and secondary metal production; powder production by spray forming; and welding, but also in more specialized uses like self-repair broken circuits that employ micro-capsules filled with liquid metals. Depending on which aspect of the process is modeled, there is a need for viscosity and density data for the relevant alloys. Historically, there are wide discrepancies in the viscosity data reported for the metallic elements and alloys.[1] For example, there is a spread of about 400% in the reported values of the viscosity for molten aluminum and about 100% for molten iron. For these reasons, 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) to critically evaluate the density and the viscosity of selected liquid metals. Thus:

(i) In 2006, recommended values for the density and the viscosity of liquid aluminum and iron were published,[2] as a result of a project supported by IUPAC.
(ii) Following this, in 2010, values for the density and viscosity for liquid copper and tin were proposed.[3] That work was also carried out under the auspices of IATP and was supported by IUPAC.
(iii) In 2011, the work was continued and reference correlations of the density and viscosity of liquid bismuth, nickel, lead, silver, and antimony were proposed.[4]
(iv) The current paper concludes the work on the density and viscosity of pure liquid metals by presenting reference correlations for liquid cadmium, cobalt, gallium, indium, mercury, silicon, thallium, and zinc. For the remaining liquid metals, very limited literature is available.

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,[2,3] 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 datasets. These limits are determined after critical evaluation of the existing datasets.

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%.

In the case, however, of the liquid metals, it was argued that these criteria needed to be relaxed slightly, especially since the uncertainty of the measurements is 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 molten cadmium, cobalt, gallium, indium, mercury, silicon, thallium, and zinc. Methods employed include: Archimedean; pycnometric; bubble-pressure; sessile-drop; falling-drop; levitation; gamma radiation attenuation. These methods have been presented in our previous compilation[2] and will only very briefly be discussed here.

The most commonly employed technique for the measurement of the density is the Archimedean technique. According to this method, a solid sinker of known weight in air is suspended by a wire attached to the arm of a balance. When the sinker is entirely immersed in the liquid metal specimen, an apparent loss of weight is observed, arising mainly from the buoyant force exerted by the liquid metal sample. The loss of weight is simply related to the density of the liquid of immersion. Another very accurate absolute technique is the pycnometric technique, which refers to the filling of a vessel or crucible of known volume with the liquid metal. Upon freezing, the solid metal specimen is weighed at room temperature. A similar technique, based on the principle of weighing the solid, is the areometric technique.

The maximum-bubble-pressure technique is based upon the formation of a hemispherical bubble of an inert gas at the tip of a capillary tube immersed to a certain depth in the melt. The density can be determined by measuring the difference in the overpressure required to form a hemispherical bubble of the inert gas at the tip of the capillary at different depths in the liquid. The technique is not as accurate as the pycnometric method but allows density measurements at higher temperatures. The sessile-drop technique employs a liquid drop of known mass resting on a plate or substrate. Provided the shape of the drop is fully symmetrical, the volume of the drop, and hence its density, can accurately be calculated.

In the levitation technique, a small drop of the liquid metal can be supported by one of the three techniques: (a) aerodynamically by gas flow in a convergent/divergent nozzle; (b) electrostatically by electrically charging the drop and holding it steady using an electrical potential; or (c) by electromagnetic forces using a high-frequency coil. In the case of aerodynamic and electrostatic levitation, the drop is heated by a high-power laser but frequently the electromagnetic field is used to both levitate and heat the drop. The volume of the drop is obtained from sectional images which are frequently taken from three orthogonal directions.

The gamma radiation attenuation technique is based on the attenuation of a γ-ray beam passing through the liquid metal. The incident beam is attenuated according to the mass of the liquid metal. Finally, a fast pulse-heating technique coupled with fast photography has recently been employed for the measurement of density of liquid metals.

3.2. Data compilation

Table 1 presents the datasets found for the measurement of the density of liquid cadmium, cobalt, gallium, indium, silicon, thallium, and zinc. In this table, the purity of the sample, 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 datasets 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 Sec. 3.1. More specifically, following the brief presentation of the various techniques employed for the measurement of the density of the liquid metals, the following can be noted:

(i) *Cadmium*: Seven investigators reported density measurements for cadmium. The measurements of Crawley[9] were performed in absolute pycnometers with low uncertainty and were considered as primary data. The measurements of Karamurzov[7] and Alchagirov *et al.*,[8] performed in an areometer densimeter, and those of Fisher and Philips,[11] taken in bubble-pressure instruments, were also part of the primary dataset, together with the γ-ray measurements of Stankus[6] and of Schneider and Heymer.[10] Finally, the measurements of Chentsov,[12] performed in a sessile-drop instrument, were considered as secondary data, as they were much higher than the results of other workers and also showed quite a different temperature gradient.
(ii) *Cobalt*: In the case of cobalt, 11 sets of density measurements were considered. All these sets were characterized by an uncertainty of less than 1%, except the measurements of Brillo *et al.*[13] and Saito *et al.*,[19] which were

TABLE 1. Datasets considered for the density of liquid cadmium, cobalt, gallium, indium, silicon, thallium, and zinc.

First author	Publ. year	Technique employed[a]	Purity (mass%)	Uncertainty quoted (%)	No. of data	Form of data[b]	Temperature range (K)
			Cadmium				
Primary data							
Stankus[6]	1992	γ-Ray (Abs)	99.99	0.2	7	P	594–700
Karamurzov[7]	1975	Areometer (Abs)	na	1.0	6	E	594–800
Alchagirov[8]	1974	Areometer (Abs)	na	0.1	6	E	594–773
Crawley[9]	1968	Pycnometer (Abs)	99.999	0.02	7	P	608–709
Schneider[10]	1956	γ-Ray (Abs)	na	0.2	5	P	658–833
Fisher[11]	1954	Bubble pressure (Abs)	99.989	na	4	P	603–673
Secondary data							
Chentsov[12]	1971	Sessile drop	99.99	1	5	E	600–800
			Cobalt				
Primary data							
Brillo[13]	2006	EML (Abs)	na	1.5	14	D	1724–1875
Sato[14]	2002	Pycnometer (Abs)	99.8	0.5	5	D	1774–1867
Stankus[6]	1992	γ-Ray (Abs)	99.9	0.5	5	P	1765–1950
Lucas[15]	1972	Archimedean (Abs)	na	0.3	5	E	1774–1972
Watanabe[16]	1971	Bubble pressure (Ads)	99.9	0.7	8	E	1793–1898
Shergin[17]	1970	Sessile drop (Abs)	na	1	7	E	1765–2123
Levin[18]	1970	Sessile drop (Abs)	na	1	10	D	1774–2077
Saito[19]	1969	EML (Abs)	na	1.4	24	D	2060–2470
Vertman[20]	1964	Sessile drop (Abs)	99.9	0.2	6	D	1769–1926
Frohberg[21]	1964	Bubble pressure (Abs)	99.97	na	6	D	1775–1843
Kirshebaum[22]	1963	Archimedean (Abs), bubble pressure (Abs)	99.9	0.2	10	D	1858–2391
Secondary data							
–							
			Gallium				
Primary data							
Yagodin[23]	2008	γ-Ray (Abs)	99.999	0.2	50	D	526–1501
Stankus[24]	1991	γ-Ray (Abs)	99.9997	0.2	15	E	310–1000
Alchagirov[25]	1974	Areometer (Abs)	na	0.1	11	E	310–773
Nal'giev[26]	1973	Pycnometer (Abs)	99.99	na	10	P	303–723
Koster[27]	1970	Pycnometer (Abs)	99.999	0.03	12	P	323–873
Nizhenko[28]	1965	Sessile drop (Abs)	na	0.03	11	E	380–1580
Secondary data							
Geng[29]	2010	Archimedean	99.9	5	20	D	312–1073
Yatsenko[30]	1972	Sessile drop (Abs)	na	1.5	18	D	327–1179
Spells[31]	1935	na	na	na	17	P	326–1373
			Indium				
Primary data							
Alchagirov[8]	2004	Pycnometer (Abs)	99.9	0.1	45	P	433–580
Wang[32]	2004	γ-Ray	na	na	14	E	429–1073
McClelland[33]	1995	Sessile drop	99.99	0.95	5	D	429–774
Stankus[24]	1991	γ-Ray (Abs)	99.9997	0.05	13	E	532–1100
Karamurzov[7]	1975	Areometer (Abs)	na	1.0	8	E	429–773
Berthou[34]	1970	Archimedean (Abs)	99.999	na	19	E	433–805
Crawley[9]	1968	Pycnometer (Abs)	99.999	0.05	12	P	434–537
Schneider[10]	1956	γ-Ray (Abs)	na	0.2	7	P	504–694
Secondary data							
Yatsenko[30]	1972	Sessile drop (Abs)	na	1.5	9	D	454–921
Williams[35]	1950	Dilatometer (Rel)	99.98	0.2	5	E	437–573
Gamertsfelder[36]	1941	Dilatometer (Rel)	na	na	5	E	430–550
			Silicon				
Primary data							
Watanabe[37]	2007	EML (Abs)	na	na	7	E	1683–1990
Zhou[38]	2003	ESL (Abs)	99.999	1.5	7	E	1683–1830
Mukai[39]	2000	Sessile drop	na	0.3	8	E	1683–1853

TABLE 1. Datasets considered for the density of liquid cadmium, cobalt, gallium, indium, silicon, thallium, and zinc.—Continued

First author	Publ. year	Technique employed[a]	Purity (mass%)	Uncertainty quoted (%)	No. of data	Form of data[b]	Temperature range (K)
Sato[40]	2000	Pycnometric (Abs)	na	0.5	7	P	1698–1853
Oshaka[41]	1997	ESL (Abs)	99.9995	0.2	7	E	1683–1825
Rhim[42]	1997	ESL (Abs)	99.9995	0.2	7	E	1683–1825
Sasaki[43]	1993	Archimedean (Abs)	na	1.1	10	D	1719–1910
Khilya[44]	1973	Sessile drop	na	1.5	7	E	1773–1863
Shergin[17]	1970	Sessile drop	na	1.0	8	E	1683–2000
Glazov[45]	1967	Archimedean (Abs)	na	1.5	7	E	1728–1906
Lucas[46]	1964	Bubble pressure (Abs)	na	0.2	6	P	1683–1923
Secondary data							
Langen[47]	1998	EML (Abs)	na	5.0	4	D	1682–1759
Vatolin[48]	1963	Pycnometer	na	2.0	1	P	1713
Logan[49]	1958	X-ray diffraction	na	1.0	1	P	1698
			Thallium				
Primary data							
Stankus[50]	1988	γ-Ray (Abs)	99.95	0.07–0.11	5	P	577–800
Kanda[51]	1979	Archimedean (Abs)	99.999	na	18	P	577–773
Martinez[52]	1973	Archimedean (Abs)	99.999	0.03	14	P	577–1178
Berthou[53]	1968	Archimedean (Abs)	99.999	na	12	E	580–1020
Crawley[54]	1968	Pycnometer (Abs)	99.999	0.1	10	P	587–781
Schneider[10]	1956	γ-Ray (Abs)	na	0.2	10	P	665–924
Secondary data							
–							
			Zinc				
Primary data							
Stankus[50]	1988	γ-Ray (Abs)	99.95	0.07–0.11	3	P	693–900
Karamurzov[7]	1975	Areometer (Abs)	na	1.0	6	E	723–953
Thresh[55]	1968	Pycnometer (Abs)	99.99	0.1	20	D	693–792
Lucas[46]	1964	Bubble pressure (Abs)	na	0.2	10	D	708–907
Gebhardt[56]	1955	Archimedean (Abs)	na	na	3	P	773–973
Secondary data							
Otter[57]	1996	Pulse-heating (Abs)	99.99	4.0	7	E	700–1300

[a]Abs = absolute; ESL = electrostatic levitation; EML = electromagnetic levitation; Rel = relative.
[b]D = diagram; E = equation; P = points.

performed by the electromagnetic levitation technique with uncertainties of 1.5% and 1.4%, respectively. Because both these sets were considered as primary data in a previous publication,[4] in this work they were also considered in the same category. All remaining datasets were also considered as primary data. The measurements of Sato *et al.*[14] were performed in an absolute pycnometer, while the measurements of Stankus[6] were performed in a γ-ray instrument. The Archimedean technique was used by Lucas[15] and Kirshenbaum and Cahill.[22] Bubble-pressure instruments were employed by Watanabe,[16] Frohberg and Weber,[21] and Kirshenbaum and Cahill.[22] Finally, Shergin,[17] Levin *et al.*,[18] and Vertman *et al.*[20] employed a sessile-drop device.

(iii) *Gallium*: In the case of gallium, the recent measurements of Geng *et al.*[29] were considered as secondary data owing to their high uncertainty. Density measurements were reported by Spells[31] in 1935, but with no details of the method or the uncertainty; these were also considered as secondary data. Also the measurements of Yatsenko *et al.*[30] performed in a sessile-drop instrument were considered as secondary data, as they showed a different temperature gradient than the other investigators. The remaining six sets of measurements were all primary data. Pycnometers were employed by Nal'giev and Ibragimov[26] and by Köster *et al.*,[27] an areometer densimeter was employed by Alchagirov,[25] while Nizhenko *et al.*[28] employed a sessile-drop instrument, and Yagodin *et al.*[23] and Stankus and Tyagel'sky[24] γ-ray instruments.

(iv) *Indium*: 11 investigators reported measurements of the density of indium. The measurements of Alchagirov *et al.*[8] and Crawley[9] were performed in absolute pycnometers and with very low uncertainty and were thus considered as primary data. The measurements of Stankus and Tyagel'sky[24] and Schneider and Heymer,[10] performed in a γ-ray instrument with very low uncertainty, were also part of the primary dataset. It should however be noted that, although the measurements of Stankus and Tyagel'sky[24] extended to 1500 K, we have not included the data above 1100 K because no other investigator performed measurements higher than 1100 K. Part of the primary dataset were also the measurements of Wang *et al.*[32] performed in a γ-ray instrument and of Karamurzov[7] performed in an areometer densimeter, as well as the measurements of McClelland and Sze[33] performed in a sessile-drop apparatus and Berthou and Tougas[53] obtained by the Archimedean technique. The measurements of Yatsenko *et al.*[30] performed in a sessile-drop instrument with 1.5% uncertainty were not included in the primary set, as they showed a different temperature gradient than the rest (the same different trend was observed in gallium). The measurements of Williams and Miller[35] and Gamertsfelder,[36] performed on a relative basis with a dilatometer, were also considered as secondary data.

(v) *Mercury*: In the case of mercury, Bigg[58] in 1964 proposed standard values for the density of mercury between −20 °C and 300 °C. The data were based on the values proposed by Beattie *et al.*[59] in 1941 and the measurements of Harlow[60] in 1913. It is worthwhile noting that both sets agreed within a few parts per million. The values proposed by Beattie were based themselves on a collection of data (Chappuis,[61] Callendar and Moss,[62] James,[63] Sears,[64] and Harlow[60]). In 1994, Sommer and Poziemski[65] published a paper on the density of mercury at 20 °C and 101 kPa after considering all recent investigators (Cook,[66] Furtig,[67] Adametz,[68] Patterson and Prowse[69]) including their own measurements. Finally in 2004, Bettin and Fehlauer[70] performed new measurements and proposed the reference values for the density of mercury that are in use today.

(vi) *Silicon*: In this case 11 sets of measurements were considered as primary data. The measurements of Watanabe *et al.*,[37] Zhou *et al.*,[38] Oshaka *et al.*,[41] and Rhim *et al.*[42] were performed in an electrostatic levitation instrument. Mukai and Yuan,[39] Khilya and Ivashchenko,[44] and Shergin[17] employed a sessile instrument, while Sasaki *et al.*[43] employed an instrument based on the Archimedean principle. A pycnometric apparatus was employed by Sato *et al.*,[40] while Lucas[46] performed his measurements in a bubble-pressure instrument. It should be noted that there is a relatively wide spread of values in the diagram. The measurements of Langen *et al.*,[47] performed in an electromagnetic levitator, are quoted with 5% uncertainty, and hence were considered as secondary data. The single measurement of Logan and Bond[49] performed in an x-ray diffraction apparatus was also considered as a secondary datum. Finally, the single measurement of Vatolin and Esin[48] performed with a 2% uncertainty was part of the secondary data.

(vii) *Thallium*: All six sets of density measurements were considered as primary data. The measurements of Stankus and Khairulin[50] and Schneider *et al.*[10] were performed in an absolute γ-ray instrument. The Archimedean technique was employed in an absolute way by Kanda and Dominique,[51] Martinez and Walls,[52] and Berthou and Tougas.[53] Finally, the measurements of Crawley[54] were obtained in an absolutepycnometer.

(viii) *Zinc*: The primary dataset is composed of five sets of measurements. The measurements of Stankus and Khairulin[50] were performed in a γ-ray instrument in an absolute way. A bubble-pressure instrument was employed by Lucas[46] in an absolute fashion. Thresh[55] employed an absolute pycnometer, Karamurzov[7] employed the areometric technique, and Gebhardt *et al.*[56] employed the Archimedean technique. Otter *et al.*[57] employed the pulse-heating technique for the measurement of liquid zinc with an uncertainty of 4%. These measurements deviated very much from all other sets and were thus considered as secondary data.

3.3. Density reference correlation

The primary density data for liquid metals, 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),

$$\rho = c_1 - c_2(T - T_{\text{ref}}), \tag{1}$$

and the coefficients c_1 (kg m^{-3}), c_2 (kg m^{-3} K^{-1}), as well as the melting temperature T_{ref} (K), are shown for each liquid metal in Table 2. In the same table, the percentage deviation (2σ) of

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})	T_{ref} (K)		Deviation (2σ) (%)
Cadmium	594–833	8008	1.251	594.219	(Ref. 71)	0.6
Cobalt	1768–2500	7827	0.936	1768.0	(Ref. 72)	2.1
Gallium	303–1500	6077	0.611	302.914	(Ref. 73)	0.4
Indium	430–1100	7022	0.762	429.748	(Ref. 73)	0.5
Silicon	1687–2000	2550	0.264	1687.0	(Ref. 41)	2.2
Thallium	576–1200	11233	1.200	576.7	(Ref. 74)	0.9
Zinc	692–910	6559	0.884	692.677	(Ref. 73)	0.7

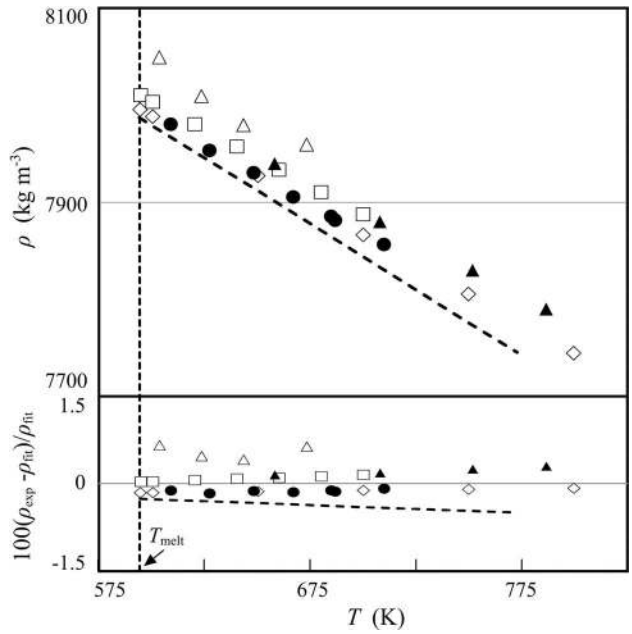

FIG. 1. Primary density data and their percentage deviations from Eq. (1) for liquid cadmium as a function of temperature. Stankus[6] (□), Karamurzov[7] (◊), Alchagirov *et al.*[8] (- -), Crawley[9] (●), Schneider and Heymer[10] (▲), Fisher and Philips[11] (Δ).

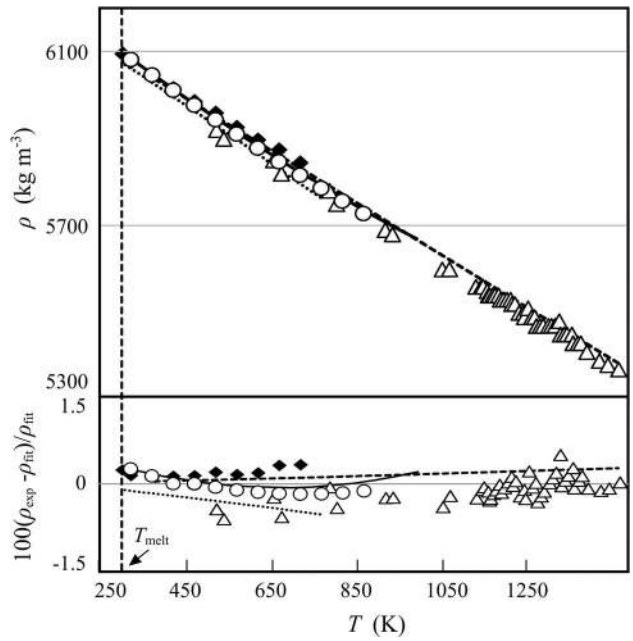

FIG. 3. Primary density data and their percentage deviations from Eq. (1) for liquid gallium as a function of temperature. Yagodin *et al.*[23] (Δ), Stankus and Tyagel'sky[24] (——), Alchagirov[25] (···), Nal'giev and Ibragimov[26] (♦), Nizhenko *et al.*[28] (- -), Köster *et al.*[27] (○).

each equation at the 95% confidence level is also shown. It should be noted, as already discussed, that in the case of mercury, since reference values do exist, no further work was done.

Figures 1–7 show the primary data and their percentage deviations from the above equation for each liquid metal, except mercury. The dashed vertical line shows the melting point for each metal. The following can be observed:

(i) In the case of cadmium (Fig. 1), gallium (Fig. 3), indium (Fig. 4), thallium (Fig. 6), and zinc (Fig. 7), the deviations from Eq. (1) are in general within the quoted uncertainty of each investigator. These six reference density correlations can be considered to represent the data well and the overall uncertainty is commensurate with the authors' claim.

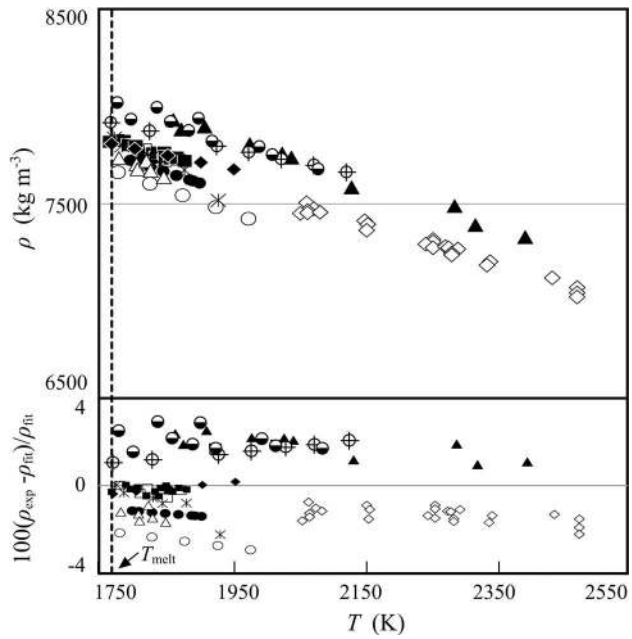

FIG. 2. Primary density data and their percentage deviations from Eq. (1) for liquid cobalt as a function of temperature. Brillo *et al.*[13] (■), Sato *et al.*[14] (□), Stankus[6] (♦), Lucas[15](○), Watanabe[16] (●), Levin *et al.*[18] (◒), Shergin[17] (⊕), Saito *et al.*[19] (◊), Frohberg and Weber[21] (Δ), Vertman *et al.*[20] (∗), Kirshenbaum and Cahill[22] (▲).

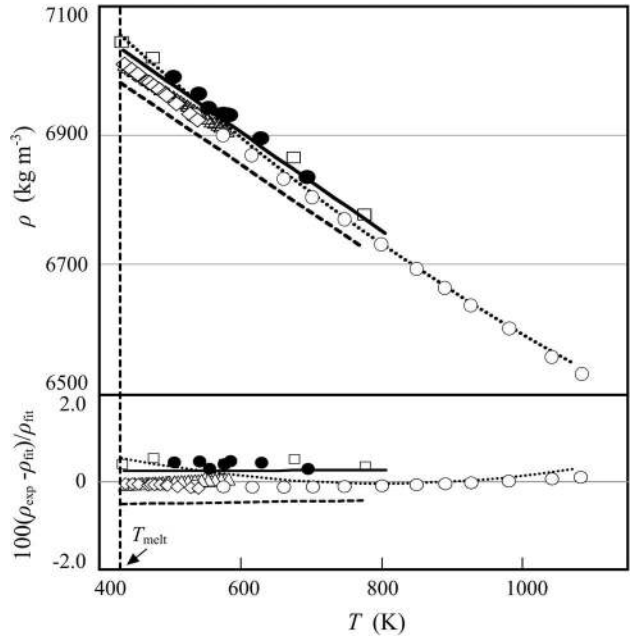

FIG. 4. Primary density data and their percentage deviations from Eq. (1) for liquid indium as a function of temperature. Alchagirov *et al.*[8] (Δ), Wang *et al.*[32] (···), Schneider and Heymer[10] (●), McClelland and Sze[33] (□), Stankus and Tyagel'sky[24] (○), Karamurzov[7] (- -), Berthou and Tougas[53] (——), Crawley[9] (◊).

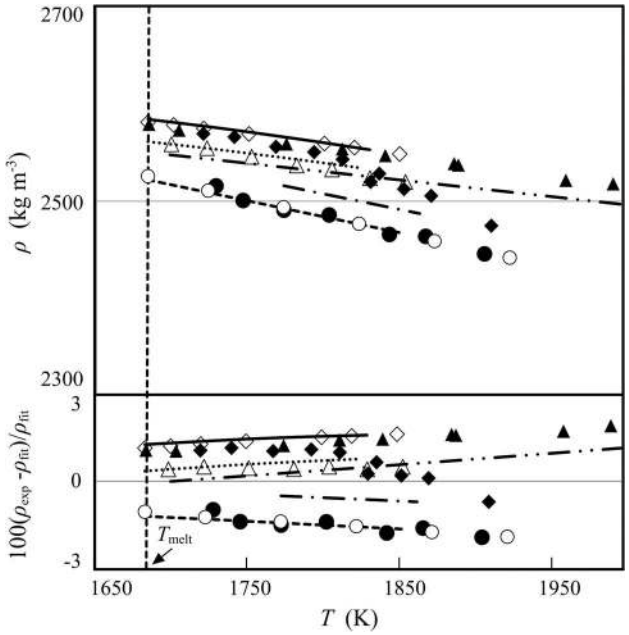

FIG. 5. Primary density data and their percentage deviations from Eq. (1) for liquid silicon as a function of temperature. Watanabe *et al.*[37] (▲), Zhou *et al.*[38] (——), Sato *et al.*[40] (Δ), Mukai and Yuan[39] (- -), Oshaka *et al.*[41] (◊), Rhim *et al.*[42] (···), Khilya and Ivashchenko[44] (-·-), Shergin[17] (-···-), Glazov *et al.*[45] (●), Sasaki *et al.*[43] (▲), Lucas[46] (○).

(ii) The deviations of the results of the measurements of the density of cobalt (Fig. 2) from Eq. (1) far exceed the quoted uncertainty of each investigator, which extend from 0.2% to 1.5%. This picture does not change, even if we restrict the primary data only to measurements of very low stated uncertainty.

(iii) A very similar picture is observed in the case of silicon (Fig. 5). Here also, the deviations from Eq. (1) far exceed the uncertainty of each investigator. It is not possible for us to resolve these discrepancies, so the correlations have an uncertainty larger than that claimed by individual authors. In the case of silicon, this might be attributed to the reactivity of silicon, because a similar observation has been made during its viscosity measurement.

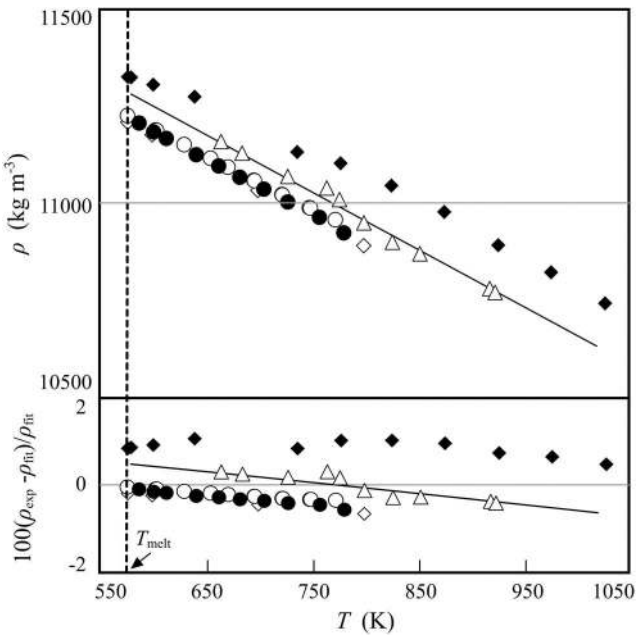

FIG. 6. Primary density data and their percentage deviations from Eq. (1) for liquid thallium as a function of temperature. Stankus and Khairulin[50] (◊), Kanda and Dominique[51] (○), Martinez and Walls[52] (♦), Berthou and Tougas[53] (——), Crawley[54] (●), Schneider *et al.*[10] (Δ).

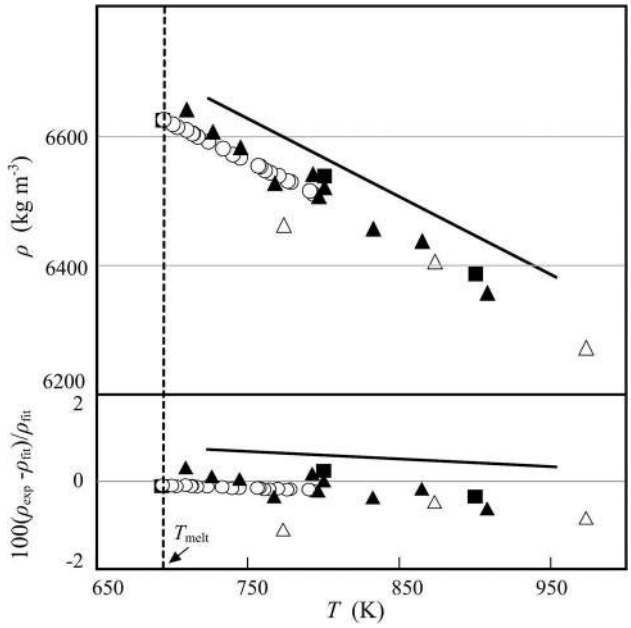

FIG. 7. Primary density data and their percentage deviations from Eq. (1) for liquid zinc as a function of temperature. Stankus and Khairulin[50] (■), Karamurzov[7] (——), Thresh[55] (○), Lucas[46] (▲), Gebhardt *et al.*[56] (Δ).

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

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. Proposed methods include: capillary; oscillating cup; rotational bob; oscillating plate; draining vessel; levitated drop, and acoustic methods. These methods have been presented in our previous compilation[2] and will not be discussed here.

Most measurements use some form of oscillating-cup viscometer. A vessel, normally a cylinder, containing the test liquid is suspended by a torsion wire and is set in motion about the vertical axis. The oscillatory motion is damped by viscous friction within the liquid, and consequently, the viscosity is determined from the decrement and time period of the motion. This method is applicable up to temperatures of ~2000 K (Ref. 75) and has a sufficiently high sensitivity for viscosities down to 1 mPa s. Unfortunately, the working equation is implicit and must be solved numerically. It should be emphasized that, according to our previous work,[2] datasets employing the equation of Knappwost[76] have not been considered as primary datasets; only those employing equations

TABLE 3. Recommended values for the density and viscosity of cadmium, cobalt, gallium, indium, mercury, silicon, thallium, and zinc.

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)	T (K)	ρ (kg m^{-3})	η (mPa s)
Liquid cadmium			Liquid cobalt			Liquid gallium			Liquid indium		
600	8001	2.708	1800	7797	4.543	350	6048	1.369	450	7007	1.748
650	7938	2.326	1850	7750	4.123	400	6018	1.158	500	6968	1.521
700	7876	2.043	1900	7703	3.761	450	5987	1.016	550	6930	1.357
750	7813	1.825	1950	7657	3.446	500	5957	0.915	600	6892	1.234
800	7751	1.654	2000	7610	3.172	550	5926	0.840	650	6854	1.139
850	7688	1.516	2050	7563	2.932	600	5895	0.783	700	6816	1.063
900	7625	1.403	2100	7516	2.719	650	5865	0.737	750	6778	1.001
			2150	7469		700	5834	0.700	800	6740	0.951
			2200	7423		750	5804	0.669	850	6702	0.908
			2250	7376		800	5773	0.643	900	6664	0.871
			2300	7329		850	5743		950	6626	0.840
			2350	7282		900	5712		1000	6587	0.813
			2400	7235		950	5682		1050	6549	
			2450	7189		1000	5651		1100	6511	
			2500	7142		1050	5621				
						1100	5590				
						1150	5559				
						1200	5529				
						1250	5498				
						1300	5468				
						1350	5437				
						1400	5407				
						1450	5376				
						1500	5346				
Liquid mercury			Liquid silicon			Liquid thallium			Liquid zinc		
250	…	1.875	1700	2547	0.605	600	11205	2.434	700	6553	3.737
300	…	1.531	1750	2533	0.571	650	11145	2.155	750	6508	3.254
350	…	1.324	1800	2520	0.541	700	11085	1.941	800	6464	2.883
400	…	1.187	1850	2507	0.514	750	11025	1.773	850	6420	2.591
450	…	1.091	1900	2494	0.490	800	10965	1.638	900	6376	2.356
500	…	1.020	1950	2481		850	10905		950	6332	2.164
550	…	0.965	2000	2467		900	10845		1000		2.005
600	…	0.921				950	10785		1050		1.871
						1000	10725		1100		1.756
						1050	10665				
						1100	10605				
						1150	10545				
						1200	10485				

based upon the work of Roscoe,[77] published in 1958, have been considered.

In addition to the oscillating-cup technique, the capillary technique[2] (and the double-capillary technique), has successfully been employed for the measurement of the viscosity of liquid metals. The capillary rheometer is generally thought to be the best method for the measurement of the viscosity of liquids,[1] and is based upon the time for a finite volume of liquid to flow through a narrow-bore tube under a given pressure. The relation between viscosity and efflux time is given by a modified Poiseuille equation or a Hagen-Poiseuille equation.[2] This technique is often used as a relative, rather than absolute, method, because the experimental procedures are simple, and any errors arising from the measurement of dimensions are thereby avoided. Measurements performed by the capillary technique are usually considered as primary data.

The electrostatic levitation (ESL) and electromagnetic levitation (EML) techniques employed for the measurement of the density of liquid metals are employed as well for the measurement of the viscosity. When employed properly, both techniques can produce very good results.

A few other measurements were performed by various secondary techniques;[2] the oscillating-sphere technique, the rotating-cylinder method, and the vibration technique are considered to produce secondary data as they do not satisfy most of the aforementioned criteria, the most important of which being the lack of a complete theory describing these techniques.

4.2. Data compilation

Table 4 presents the datasets found for the measurement of the viscosity of liquid cadmium, cobalt, gallium, indium, mercury, silicon, thallium, and zinc. As in the case of the density measurements, papers prior to 1930 were not considered, because sample purity was disputed before that time. In

TABLE 4. Datasets considered for the viscosity of liquid cadmium, cobalt, gallium, indium, mercury, silicon, thallium, and zinc.

First author	Publ. year	Technique employed[a]	Purity (mass%)	Uncertainty quoted (%)	No. of data	Form of data[b]	Temperature range (K)
			Cadmium				
Primary data							
Djemili[78]	1981	Oscillating cup (Abs)	99.999	na	12	P	598–710
Iida[79]	1980	Oscillating cup (Rel)	99.99	na	5	P	607–804
Iida[80]	1975	Capillary (Abs)	99.9999	0.5	6	P	613–873
Kanda[81]	1973	Oscillating cup (Abs)	99.999	1.0	1	D	623
Crawley[82]	1969	Oscillating cup (Abs)	99.999	0.5	10	P	595–724
Menz[83]	1966	Double capillary (Abs)	99.999	2.0	3	D	606–692
Secondary data							
Fisher[11]	1954	Oscillating cup	na	na	5	P	623–723
			Cobalt				
Primary data							
Sato[84]	2005	Oscillating cup (Abs)	99.9	1.0	20	P	1755–1881
Lad'yanov[85]	2000	Oscillating cup (Abs)	na	1.5	8	E	1773–1973
Kaplun[86]	1977	Oscillating cup (Rel)	na	5.0	9	E	1797–2090
Watanabe[16]	1971	Oscillating cup (Rel)	na	5.0	31	D	1781–2032
Cavalier[87]	1963	Oscillating cup (Abs)	99.87	na	8	P	1723–2023
Secondary data							
Paradis[88]	2008	ESL (Abs)	99.9	na	11	E	1690–1950
Han[89]	2002	EML (Abs)	99.999	na	6	D	1772–1973
Bodakin[90]	1978	Oscillating cup	na	3.0	8	D	1759–1972
			Gallium				
Primary data							
Tippelskirch[91]	1976	Oscillating cup (Abs)	99.99	0.5	42	P	307–800
Genrikh[92]	1972	Vibration method (Rel)	99.9	1.5	33	P	337–366
Menz[83]	1966	Double capillary (Abs)	na	2.0	4	D	449–602
Secondary data							
Iida[79]	1980	Oscillating cup (Rel)	99.99	na	6	P	293–1293
Iida[80]	1975	Capillary (Abs)	99.99	0.5	14	P	305–547
Spells[31]	1935	Capillary (Rel)	na	na	17	P	326–1373
			Indium				
Primary data							
Walsdorfer[93]	1988	Capillary (Abs)	na	na	12	P	443–1273
Djemili[78]	1981	Oscillating cup (Abs)	99.999	na	16	P	436–899
Iida[80]	1975	Capillary (Abs)	99.99	0.5	12	P	443–1273
Ganovici[94]	1969	Oscillating cup	99.999	1.0	7	P	438–1073
Crawley[82]	1969	Oscillating cup (Abs)	na	0.5	23	P	432–607
Secondary data							
Cheng[95]	2003	X-ray	99.999	5	12	D	561–1023
Nakajima[96]	1976	Oscillating cup (Rel)	na	…	20	D	453–653
Culpin[97]	1957	Oscillating sphere (Rel)	na	…	8	P	437–607
			Mercury				
Primary data							
Grouvel[98]	1977	Oscillating cup (Abs)	na	1.5	12	P	293–450

TABLE 4. Datasets considered for the viscosity of liquid cadmium, cobalt, gallium, indium, mercury, silicon, thallium, and zinc.—Continued

First author	Publ. year	Technique employed[a]	Purity (mass%)	Uncertainty quoted (%)	No. of data	Form of data[b]	Temperature range (K)
Iida[99]	1973	Capillary (Abs)	99.999	0.5	30	P	235–513
Menz[83]	1966	Double capillary (Abs)	99.999	2.0	1	D	461
Thresh[100]	1965	Oscillating cup (Abs)	99.98	1.0	5	P	296–373
Suhrmann[101]	1955	Capillary	na	0.5	18	P	234–303
Chalilov[102]	1938	Capillary	na	na	34	P	298–833
Secondary data							
–							
			Silicon				
Primary data							
Zhou[38]	2003	ESL (Abs)	99.999	7.0	11	D	1634–1844
Nishimura[103]	2002	Oscillating cup (Abs)	na	na	8	D	1826–1721
Sato[14]	2002	Oscillating cup (Abs)	na	3.0	27	P	1664–1790
Sasaki[104]	1995	Oscillating cup (Abs)	na	na	25	D	1685–1883
Secondary data							
Rhim[105]	2000	ESL (Abs)	na	10	9	D	1589–1754
Kakimoto[106]	1989	Oscillating cup (Abs)	na	na	12	D	1691–1871
			Thallium				
Primary data							
Walsdorfer[107]	1988	Capillary (Abs)	na	na	3	P	623–723
Kanda[51]	1979	Oscillating cup (Abs)	99.999	1.0	18	P	576–773
Crawley[54]	1968	Oscillating cup (Abs)	99.999	0.5	11	P	576–730
Cahill[108]	1965	Oscillating cup (Rel)	99.99	2	10	P	644–800
Secondary data							
Andrianova[109]	1971	Oscillating cup (Abs)	na	3.3	8	E	500–1200
			Zinc				
Primary data							
Mudry[110]	2008	Oscillating cup (Abs)	99.99	3.0	16	D	766–936
Iida[79]	1980	Oscillating cup (Rel)	99.9	4.0	5	P	700–913
Iida[80]	1975	Capillary (Abs)	99.98	0.5	6	P	698–973
Harding[111]	1975	Oscillating cup	99.99	1.2	12	D	676–809
Thresh[100]	1965	Oscillating cup (Abs)	99.99	1.0	36	P	695–744
Ofte[112]	1963	Oscillating cup (Abs)	99.99+	na	14	P	693–1096
Gebhardt[56]	1955	Oscillating cup (Abs)	na	na	3	P	500–700
Secondary data							
Jeyakumar[113]	2011	Rotated cylinder	99.99	5.0	4	D	697–771
Yao[114]	1952	Oscillating cup (Rel)	99.9962	na	38	P	692–873
Hopkins[115]	1950	Oscillating cup	99.83	na	6	P	702–753

[a]Abs = absolute; ESL = electrostatic levitation; EML = electromagnetic levitation; Rel = relative.
[b]D = diagram; E = equation; P = points.

the table, for every dataset, the technique employed, the purity of the sample, the uncertainty quoted, the form of the data presented, the number of data points as well as the temperature range they refer, are also shown. The datasets have been classified into primary and secondary sets according to the criteria presented in Sec. 2 and in conjunction with the techniques described in Sec. 4.1.

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

(i) *Cadmium*: The primary dataset is composed of six sets of measurements. The oscillating-cup technique was employed successfully by Djemili *et al.*,[78] Iida *et al.*,[79] Kanda and Falkiewicz,[81] and Crawley and Thresh,[82] while capillary viscometers were employed by Iida *et al.*[80] and Menz and Sauerwald.[83] The measurements of Fisher and Phillips,[11] performed by the oscillating-cup technique, were considered as secondary, as according to our previous work[2] datasets employing the equation of Knappwost[76] have not been considered as primary datasets.

(ii) *Cobalt*: Five datasets, Sato *et al.*,[84] Lad'yanov *et al.*,[85] Kaplun and Avaliani,[86] Watanabe,[16] and Cavalier,[87] all employing the oscillating-cup technique, composed the primary dataset. The recent measurements of Paradis *et al.*[88] performed by the ESL were not considered as primary data as they were much higher than everybody else. The electromagnetic EML was employed by Han *et al.*;[89] these measurements showed a distinctively different slope with temperature than the rest of the data and were thus considered as secondary. Finally, the measurements of Bodakin *et al.*[90] were also considered as secondary because they were only presented in a very small diagram.

(iii) *Gallium*: There are six sets of measurements of the viscosity of gallium. The measurements of Spells[31] performed in 1935 in a relative basis were considered as secondary data. Also, the measurements of Iida[79] are much higher than the rest and were thus considered also as part of the secondary data. The remaining three sets formed the primary data. The measurements of Tippelskirch,[91] performed in an absolute oscillating cup with an uncertainty of 0.5%, are probably the best measurements. They covered a range from 307 to 1806 K, but since no other investigator performed measurements over 800 K, they were restricted to this temperature. Also part of the primary sets were the measurements of Genrikh *et al.*[92] and Menz and Sauerwald.[83]

(iv) *Indium*: In the case of indium, the primary data are composed from five datasets: the measurements of Djemili *et al.*,[78] Ganovici and Ganovici,[94] and Crawley and Thresh[82] were performed in oscillating cup instruments, while the measurements of Walsdorfer *et al.*[93] and Iida *et al.*[80] were performed in capillary viscometers. The data of Cheng *et al.*,[95] performed by the x-ray diffraction technique in a relative manner, as well as the data of Culpin,[97] performed in an oscillating-sphere instrument, were not considered as primary data, since these techniques were never fully developed. The data of Nakajima[96] were also considered as secondary data according to our aforementioned discussion, because the equation of Knappwost was employed in the interpretation of the oscillating-cup measurements.

(v) *Mercury*: All six datasets were considered as primary data. The oscillating-cup technique was employed by Grouvel *et al.*[98] and Thresh,[100] while Iida *et al.*,[99] Menz and Sauerwald,[83] Suhrmann and Winter,[101] and Chalilov[102] employed the capillary technique.

(vi) *Thallium*: There are five sets of viscosity measurements. The measurements of Kanda and Dominique[51] and of Crawley[54] were performed in an oscillating-cup viscometer in an absolute way and were part of the primary dataset. The measurements of Cahill and Grosse[108] were obtained in an oscillating-cup viscometer but in a relative way. This set was also part of the primary data. The measurements of Walsdorfer *et al.*[107] were obtained in an absolute capillary instrument, and were also considered as primary data. Finally, the diffusivity measurements of Andrianova *et al.*[109] were considered as secondary data.

(vii) *Silicon*: Sato *et al.*[14] performed experiments with the oscillating-cup technique, employing cups made from different materials (Al_2O_3, Si_3N_4, PBN (pyrolytic boron nitride), SiO_2, 8% YSZ-yittria stabilized zirconia, SiC, and graphite). They concluded that all of the above materials produced excellent results, except the cups made from SiC and graphite which produced very high viscosity values. Sato *et al.* concluded by stating that the reasons for this difference were not entirely clear, but were related to the wettability of the material. Sasaki *et al.*[104] employed two different cups made from PBN and SiC. Consistent with the analysis of Sato *et al.*,[14] the values obtained with the SiC cup were too high; hence only the PBN-cup measurements were considered as primary data. Nishimura *et al.*[103] employed a SiC cup, but their viscosity values were very low, near the values of Sato *et al.* They argued that this was attributed to the very large inertia disk that they employed. These measurements were also considered as primary data. Zhou *et al.*[38] employed an upgraded ESL, trying to take care of all fine corrections. His measurements also formed part of the primary data. Finally, Rhim and Ohsaka[105] employed the first version of the ESL, and their data were considered as secondary data together with the data of Kakimoto *et al.*[106] whose measurements were only presented in a very small diagram.

(viii) *Zinc*: The primary data are composed of seven sets of viscosity measurements. Six of them, Mudry *et al.*,[110] Iida *et al.*,[79] Harding and Davis,[111] Thresh,[100] Ofte and Wittenberg,[112] and Gebhardt *et al.*[56] were performed in oscillating-cup instruments. Iida *et al.*[80] also performed viscosity measurements with a capillary viscometer. The measurements of Jeyakumar *et al.*,[113] performed in a concentric-cylinder relative instrument, were considered as secondary data together with the data of Hopkins and Toye[115] and Yao and Kondig,[114] which were both performed in oscillating-cup instruments but employed Knappwost's equation for the analysis of the data.

4.3. Viscosity reference correlation

The primary viscosity data for liquid cadmium, cobalt, gallium, indium, mercury, silicon, thallium, and zinc, shown in Table 4, were employed in a regression analysis as a function of the temperature. The data were weighted according

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) (%)
Cadmium	900–1300	0.4239	513.89	9.4
Cobalt	1768–2100	0.9030	2808.7	14.0
Gallium	304–800	0.4465	204.03	13.5
Mercury	234–600	0.2561	132.29	2.1
Indium	429–1000	0.3621	272.06	7.3
Silicon	1685–1900	1.0881	1478.7	15.7
Thallium	577–800	0.3017	412.84	5.1
Zinc	695–1100	0.3291	631.12	9.3

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),

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

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

Figures 8–15 show the primary viscosity data and their percentage deviations from the above equation for each liquid metal. The dashed vertical line shows the melting point for each metal. The following can be observed:

(i) In the case of mercury (Fig. 11) and thallium (Fig. 14), the deviations from Eq. (2) are in general within the quoted uncertainty of each investigator. These two reference viscosity correlations can be considered very good.

(ii) In the case of cadmium (Fig. 8), indium (Fig. 12), and zinc (Fig. 15), the deviations from Eq. (2) are somewhat larger. Nevertheless, these are also acceptable correlations.

(iii) Finally, in the case of cobalt (Fig. 9), gallium (Fig. 10), and silicon (Fig. 13), the deviations from Eq. (2) are

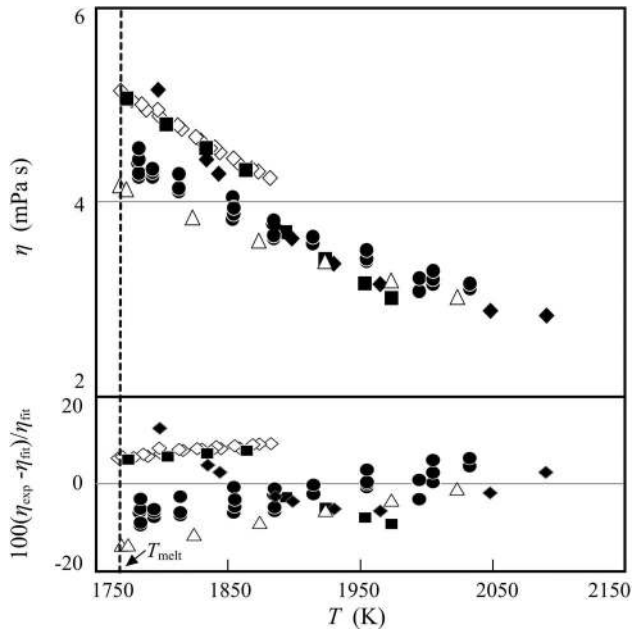

FIG. 9. Primary viscosity data and their percentage deviations from Eq. (2) for liquid cobalt as a function of temperature. Sato *et al.*[84] (◊), Lad'yanov *et al.*[85] (■), Watanabe[16] (●), Kaplun and Avaliani[86] (♦), Cavalier[87] (Δ).

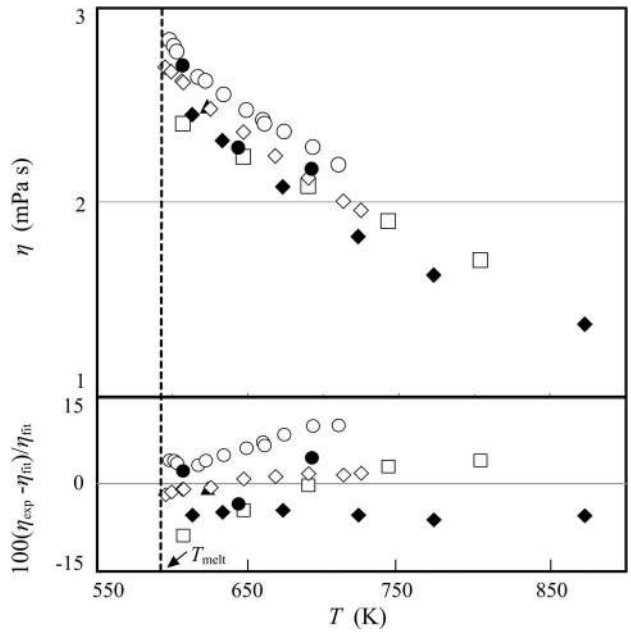

FIG. 8. Primary viscosity data and their percentage deviations from Eq. (2) for liquid cadmium as a function of temperature. Djemili *et al.*[78] (○), Iida *et al.*[79] (□), Iida *et al.*[80] (♦), Kanda and Falkiewicz[81] (▲), Crawley and Thresh[82] (◊), Menz and Sauerwald[83] (●).

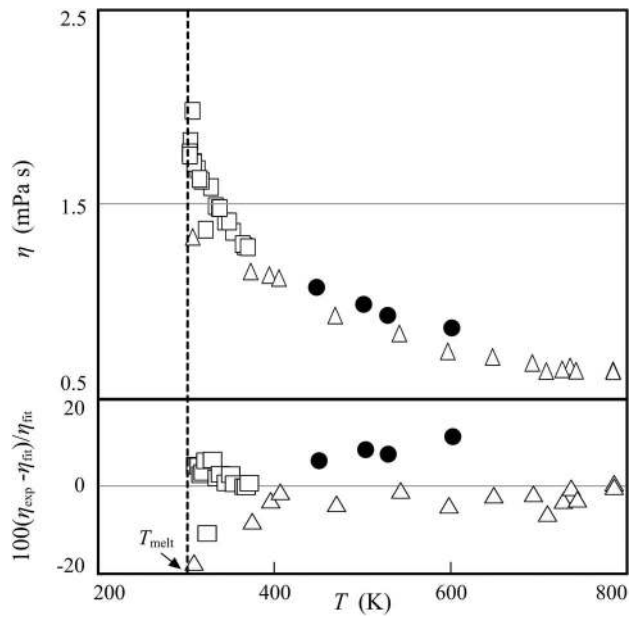

FIG. 10. Primary viscosity data and their percentage deviations from Eq. (2) for liquid gallium as a function of temperature. Tippelskirch[91] (Δ), Genrikh *et al.*[92] (□), Menz and Sauerwald[83] (●).

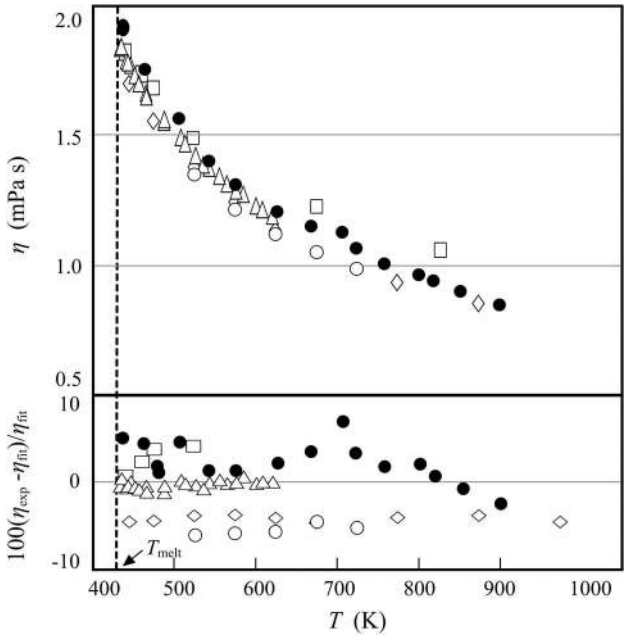

FIG. 11. Primary viscosity data and their percentage deviations from Eq. (2) for liquid indium as a function of temperature. Walsdorfer *et al.*[93] (○), Djemili *et al.*[78] (●), Iida *et al.*[80] (◊), Ganovici and Ganovici[94] (□), Crawley and Thresh[82] (Δ).

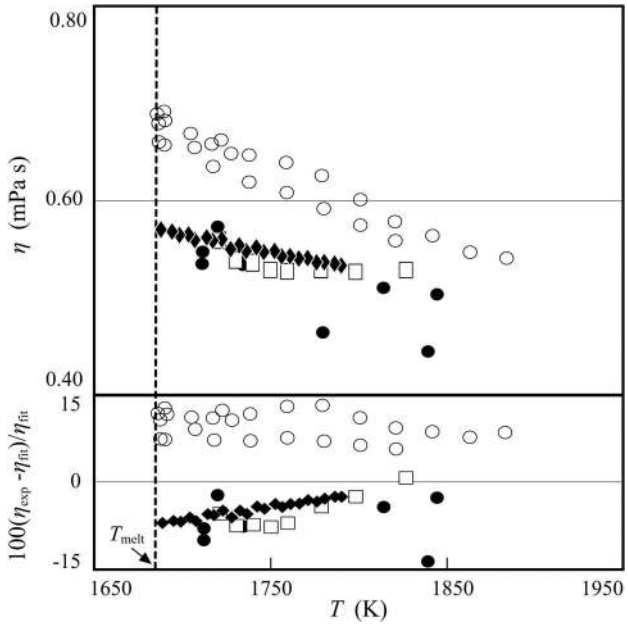

FIG. 13. Primary viscosity data and their percentage deviations from Eq. (2) for liquid silicon as a function of temperature. Zhou *et al.*[38] (●), Nishimura *et al.*[103] (□), Sato *et al.*[14] (♦), Sasaki *et al.*[104] (○).

quite high. This is attributed to the discrepancies between the various authors, probably arising from the difficulties associated with the measurement of the viscosity of these three liquid metals – certainly that was the case with silicon. These three correlations are the best that can be achieved with the sets of measurements presently available.

Viscosity values calculated from Eq. (2) are contained in Table 3.

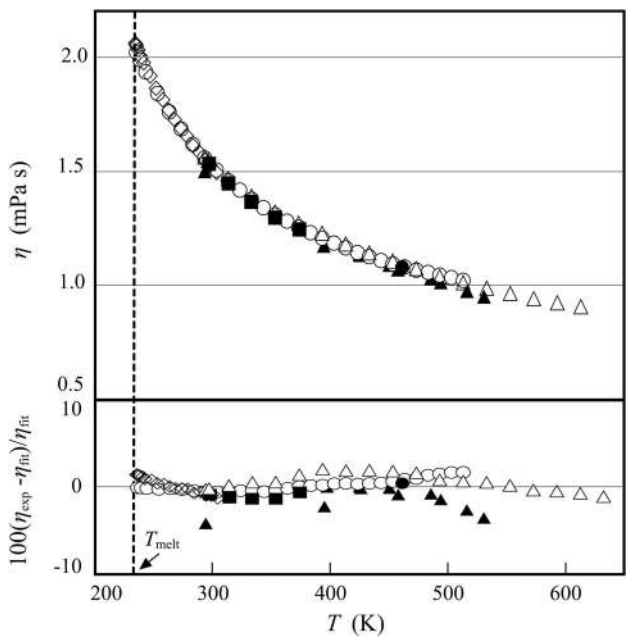

FIG. 12. Primary viscosity data and their percentage deviations from Eq. (2) for liquid mercury as a function of temperature. Grouvel *et al.*[98] (▲), Iida *et al.*[99] (○), Menz and Sauerwald[83] (●), Thresh[100] (■), Suhrmann and Winter[101] (◊), Chalilov[102] (Δ).

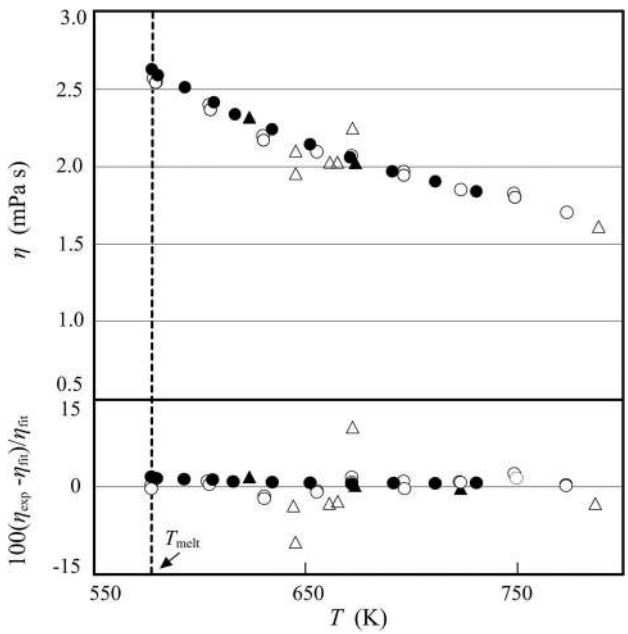

FIG. 14. Primary viscosity data and their percentage deviations from Eq. (2) for liquid thallium as a function of temperature. Walsdorfer *et al.*[93] (▲), Kanda and Dominique[51] (○), Cahill *et al.*[108] (Δ), Crawley[54] (●).

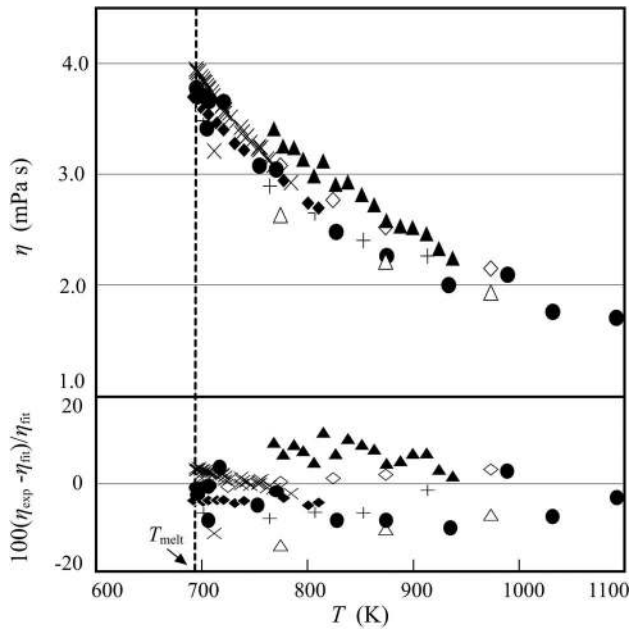

FIG. 15. Primary viscosity data and their percentage deviations from Eq. (2) for liquid zinc as a function of temperature. Mudry *et al.*[110] (▲), Iida *et al.*[79] (+), Iida *et al.*[80] (◊), Harding and Davis[111] (♦), Thresh[100] (X), Ofte and Wittenberg[112] (●), Gebhardt *et al.*[56] (Δ).

5. Conclusions

The available experimental data for the density and viscosity of liquid cadmium, cobalt, gallium, indium, mercury, silicon, thallium, and zinc 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 liquid cadmium, cobalt, gallium, indium, silicon, thallium, and zinc are, respectively, characterized by deviations of 0.6%, 2.1%, 0.4%, 0.5%, 2.2%, 0.9%, and 0.7% at the 95% confidence level. In the case of mercury, since density reference values did exist, no further work was carried out in this paper. The standard reference correlations for the viscosity of liquid cadmium, cobalt, gallium, indium, mercury, silicon, thallium, and zinc are, respectively, characterized by deviations of 9.4%, 14.0%, 13.5%, 2.1%, 7.3%, 15.7%, 5.1%, and 9.3% at the 95% confidence level.

It is apparent that more work on the measurement of the density of liquid cobalt and silicon, as well as on the measurement of the viscosity of liquid cobalt, gallium, and silicon, is still needed.

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 metals is not sufficiently high that the variation exceeds the uncertainty in the correlations reported here.

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