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Precise measurement of the densities of liquid Bi, Sn, Pb and Sb

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Abstract

The densities of liquid Bi, Sn, Pb and Sb have been precisely measured from the melting point up to about 1100 K using an improved Archimedean method. The densities at the melting point for liquid Bi, Sn, Pb and Sb are 10.042×10^3, 6.983×10^3, 10.635×10^3 and 6.454×10^3 kg m^{-3}, respectively. Comparisons between our data and those from the literature have been made and they show the present results to be more reliable. Rather than a linear fit for the temperature dependence of the density, a slight deviation from linearity in the temperature dependence of the densities has been observed.

1. Introduction

Liquid metals are in common use in modern industries/areas such as casting, metallurgy, welding, glass forming, the nuclear industry and crystal growth. In addition, how the physical properties and structures change with temperature, concentration and pressure is also of interest to researchers in physics. Density is a basic parameter in discussing the nature and behaviour of liquid metals. Measuring density values is necessary to the process of computer simulation, the calculation of other physical properties and extracting quantitative structural information from diffraction spectra.

However, due to difficulties arising from the high temperature and high vapour pressure, most published density data on liquid metals either show large discrepancies between different authors' results or were measured with large temperature intervals which makes the density data less reliable [1]. Furthermore, most published density data were measured before the 1970s except a few density data re-measured in recent years, for liquid Si and Ge for instance.

The physical properties and structure of pure liquid Sn, Sb and Bi have attracted much attention in the last few decades because the static structure factors $S(q)$ for these liquid

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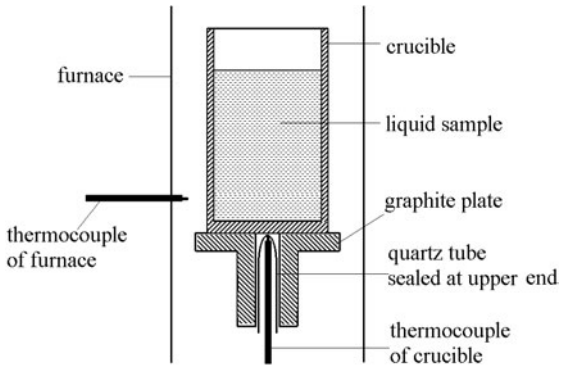

Figure 1. The schematic arrangement of the apparatus for the present measurement.

elements show a shoulder on the higher-wavevector side of the first peak [2], which cannot be account for by the random hard-spheres-packing model usually employed for normal liquid metals, such as liquid Pb. In this paper, the densities of liquid Sn, Sb, Bi and Pb from their respective melting points up to about 1110 K were measured precisely using a modified Archimedean method. The results are compared with previous data collected from the literature. More accurate values at the melting points and the temperature dependences above the melting points of the densities for these liquid metals are given.

2. Experimental details

The principle and details of the present measurement are basically the same as those described by Chen *et al* [3].

Both the crucible and the probe were made of pure graphite (99.99%), which neither reacts with nor soaks into liquid Bi, Sn, Pb and Sb. The linear thermal expansion coefficient of pure graphite is taken as 8.8×10^{-6} K^{-1} [4]. Two kinds of probe were used in present work, whose volumes are both about 4 cm^3. A two-bob probe was used to make the measured density data more stable. The probes were calibrated in advance at room temperature by using pure Hg whose density was chosen as ρ_{Hg} (293 K) $= 13.545\,85 \times 10^3$ kg m^{-3} [5]. The purities of the specimens used in the present work are Bi: 99.6%; Sn: 99.9%; Pb: 99.99%; and Sb: 99.999%.

Figure 1 shows the schematic arrangement of the apparatus for the present measurement. The temperature was measured and controlled by two Eurotherm 818P4 thermal controllers. Two Pt–10%Rh thermocouples were used in the measurements. One was located near the SiC heater, for controlling the temperature of the furnace. The other was under the crucible, for measuring the temperature of the melt, which was sealed in a quartz tube to separate it from the graphite crucible. By carefully observing the melting point of each specimen upon heating, the temperature was calibrated. The temperature distribution in the melt measured under experimental conditions indicated that the homogeneity of the temperature was within ± 4 K. In the experiment the whole system was evacuated down to about 4×10^{-3} Pa and then about 0.8 atm pure Ar (99.999%) was filled in as a protective atmosphere. All weight parameters were measured by using an AND FX-200 electronic balance with a sensitivity of 0.001 g. The relative accuracy of the density determined was better than 0.05%.

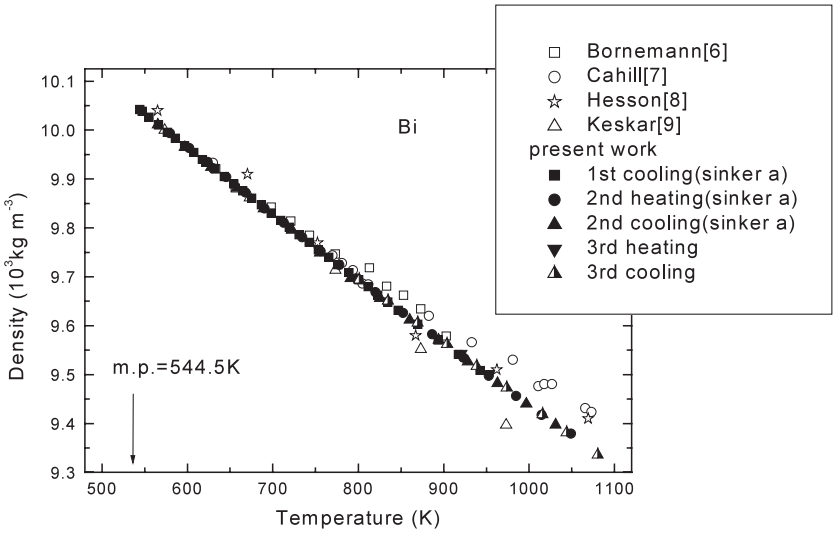

Figure 2. The density of liquid Bi at different temperatures from the present work compared with selected literature data.

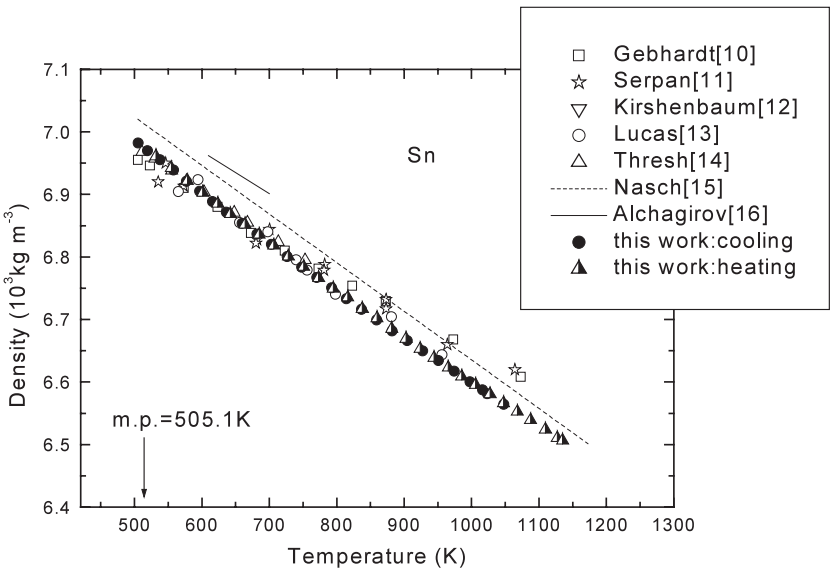

Figure 3. The density of liquid Sn at different temperatures from the present work compared with selected literature data.

3. Results and discussion

The temperature dependences of the densities of liquid Bi, Sn, Pb and Sb determined from their melting points up to about 1100 K are plotted in figures 2–5, respectively, in which the comparisons between our results and selected data from the literature are also given. The density data for these four liquid metals are spread widely in the literature; only reliable data are given in this article, for comparison.

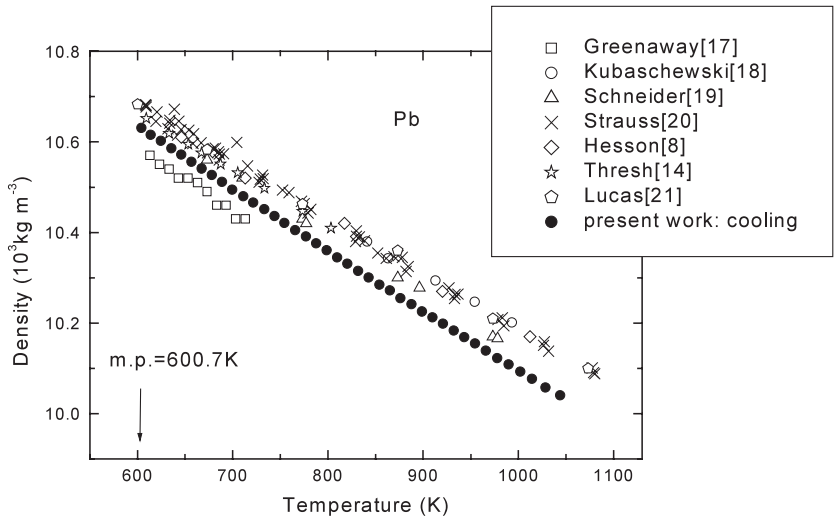

Figure 4. The density of liquid Pb at different temperatures from the present work compared with selected literature data.

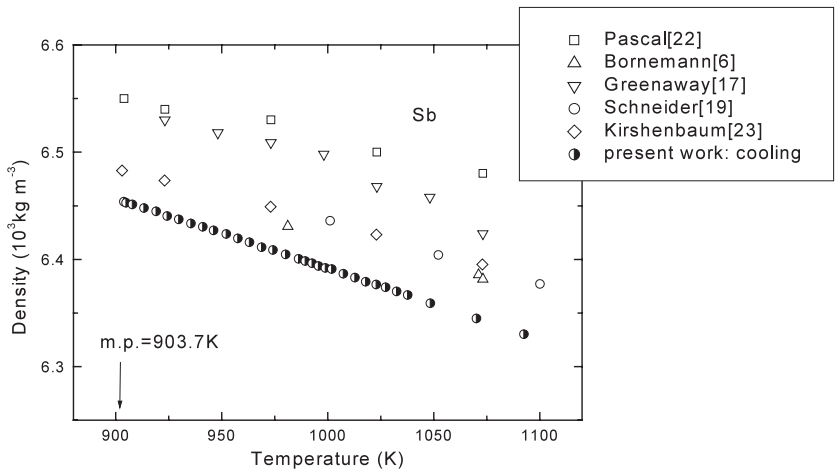

Figure 5. The density of liquid Sb at different temperatures from the present work compared with selected literature data.

3.1. Liquid bismuth

In total, five measurement cycles were made for the density of liquid Bi. Three cycles were in cooling processes and the other two in heating processes. The specimen was first heated up to a temperature much higher than its melting point (this happened to be 943 K in the first cycle) and held for 30 min at that temperature. The data points were collected with temperature intervals of about 20 or 30 K. At each destination temperature the furnace was held for 15 to 20 min to make sure the melt temperature was uniform. The cooling process ended at (or near) the melting point (544.5 K for Bi) and then the temperature was increased for the heating cycle measurement.

Figure 2 plots the measured density of liquid Bi from the melting point 545 up to 1080 K. The density decreases with temperature from 10.042×10^3 kg m^{-3} at the melting point down to 9.336×10^3 kg m^{-3} at 1080 K and can be described by a quadratic equation:

$$\rho(\mathrm{Bi}) = \rho_m - 14.9 \times 10^{-4} \times (T - 544.5) + 1.68 \times 10^{-7} \times (T - 544.5)^2 \quad (1)$$

where $\rho_m = 10.042 \times 10^3$ kg m^{-3} is the liquid density at the melting point.

Figure 2 indicates that the density values obtained during the cooling processes agree well with those obtained during heating processes. In last two processes a modified two-bob probe was used and this produced the same density values as the one-bob probe; however, the two-bob probe made the balance more stable for recording. The density data on liquid Bi selected from the literature [6–9] are also shown in figure 2.

3.2. Liquid tin

For liquid Sn one cooling process and one heating process were carried out from 506 K up to 1135 K. In this temperature range the density decreases from 6.982×10^3 kg m^{-3} to 6.507×10^3 kg m^{-3} and can be described by a quadratic equation:

$$\rho(\mathrm{Sn}) = \rho_m - 9.18 \times 10^{-4} \times (T - 504.4) + 1.49 \times 10^{-7} \times (T - 504.4)^2 \quad (2)$$

where $\rho_m = 6.983 \times 10^3$ kg m^{-3} is the liquid density at the melting point. The density data obtained in this work are plotted in figure 3 and compared with some selected literature data [10–16]. The density values in the heating and cooling processes for liquid Sn agree well with each other. The density of liquid Sn measured with a gamma-ray attenuation method by Nasch [15] was of rather poor accuracy. The data from Alchagirov [16] lay in too narrow a temperature range and the values were much higher than others. Kirshenbaum [12] measured the density of liquid Sn from the melting point up to the boiling point, 2753 K, by using an Archimedean method and reported a linear temperature dependence of the density. However, the density values obtained by Kirshenbaum were also much higher than other results.

3.3. Liquid lead

In view of the fact that good agreement in heating and cooling procedures was achieved in the density measurements of both liquid Bi and Sn, only a cooling process was performed for measuring the density of liquid Pb, from 1044 down to 604 K. The density decreases from 10.631×10^3 kg m^{-3} at 604 K to 10.040×10^3 kg m^{-3} at 1044 K and can be described by the following quadratic equation:

$$\rho(\mathrm{Pb}) = \rho_m - 15.5 \times 10^{-4} \times (T - 600.7) + 1.85 \times 10^{-7} \times (T - 600.7)^2 \quad (3)$$

where $\rho_m = 10.635 \times 10^3$ kg m^{-3} is the density of liquid Pb at the melting point. The density values for liquid Pb obtained in present work are shown in figure 4 in which some selected data from the literature [8, 14, 17–21] are also given. Our values are 0.5% lower than most of those from the literature and are about 0.5% higher than that determined by Greenaway in 1947 [17] with a maximum-bubble-pressure method.

3.4. Liquid antimony

The density of liquid Sb from 1093 K down to its melting point 904 K was measured only in one cooling process due to the severe evaporation. The density of liquid Sb decreases from 6.454×10^3 kg m^{-3} at the melting point 904 K to 6.330×10^3 kg m^{-3} at 1093 K and can be described by a quadratic equation:

$$\rho(\mathrm{Sb}) = \rho_m - 5.18 \times 10^{-4} \times (T - 903.7) - 0.964 \times 10^{-7} \times (T - 903.7)^2 \quad (4)$$

Table 1. Melting point density and temperature dependence of the density for Bi, Sn, Pb and Sb.

Element	ρ_m (10^3 kg m^{-3})	B (10^{-4} kg m^{-3} K^{-1})	C (10^{-7} kg m^{-3} K^{-2})
Bi	10.042	−14.9	1.68
Sn	6.983	−9.18	1.49
Pb	10.635	−15.5	1.85
Sb	6.454	−5.18	−0.964

where $\rho_m = 6.454 \times 10^3$ kg m^{-3} is the liquid density at the melting point. Figure 5 indicates that the density data for liquid Sb published in the literature [6, 17, 19, 22, 23] are much more scattered than those for liquid Bi, Sn and Pb and the present values are lower than all selected literature values.

In the temperature ranges studied, all the curves obtained for density versus temperature fitted as $\rho = \rho_m + B(T - T_m) + C(T - T_m)^2$ deviate somewhat from the well-known empirical linear equation [1] (exceptions are Al [24], Ga [25] and Si [26]). No abrupt changes can be observed in any of the density–temperature curves. The densities of liquid Bi, Sn, Pb and Sb at their melting points as well as the parameters in equations (1)–(4) are summarized in table 1.

4. Conclusions

The densities of liquid Bi, Sn, Pb and Sb were measured precisely from their melting points up to about 1100 K by using an improved Archimedean method. The density values at the melting points and the temperature dependences of the densities of these liquids are given. The comparison between our data and those from the literature shows the present results to be more systematic and reliable. The temperature dependence of the density shows a slight deviation from a linear equation.

Acknowledgments

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References

[1] Iida T and Guthrie R I L 1988 *The Physical Properties of Liquid Metals* (Oxford: Clarendon)
[2] North D M, Enderby J E and Egelstaff P A 1968 *J. Phys. C: Solid State Phys.* **1** 1075
[3] Chen X M, Wang Q and Lu K Q 1999 *J. Phys.: Condens. Matter* **11** 10335
[4] Weast R C, Lide D R, Astle M I and Beyer W H (ed) 1989 *CRC Handbook of Chemistry and Physics* 70th edn (Boca Raton, FL: Chemical Rubber Company Press)
[5] Ambrose D 1990 *Metrologia* **27** 245
[6] Bornemann K and Sauerwald F 1922 *Z. Metallk.* **14** 254
[7] Cahill J A and Kirshenbaum A D 1963 *J. Inorg. Nucl. Chem.* **25** 501
[8] Hesson J C, Shimotake H and Tralmer J M 1968 *J. Met.* **20** 6
[9] Keskar A R and Hruska S J 1970 *Metall. Trans.* **1** 2357
[10] Gebhardt E and Kustlin K 1957 *Z. Metallk.* **48** 636
[11] Serpan C Z and Witterbeng L J 1961 *Trans. Metall. Soc. AIME* **221** 1017
[12] Kirshenbaum A D and Cahill J A 1962 *Trans. ASM* **55** 845
[13] Lucas L D 1964 *Mem. Sci. Rev. Metall.* **21** 1

[14] Thresh H R, Crawley A F and White D W G 1968 *Trans. Metall. Soc. AIME* **242** 819
[15] Nasch P M and Steinemann S G 1995 *Phys. Chem. Liq.* **29** 43
[16] Alchagirov B B and Chochaeva A M 2000 *High Temp.* **38** 44
[17] Greenaway H T 1947 *J. Inst. Met.* **74** 133
[18] Kubaschewski O and Hurnle R 1951 *Z. Metallkde.* **42** 129
[19] Schneider A and Heymer G 1956 *Z. Anorg. Allg. Chem.* **286** 97
[20] Strauss S W, Richards L E and Brown B F 1960 *Nucl. Sci. Eng.* **17** 442
[21] Lucas L D 1972 *Mem. Sci. Rev. Metall.* **69** 359
[22] Pascal P 1914 *Rev. Metall.* **1** 469
[23] Kirshenbaum A D and Cahill J A 1962 *Trans. ASM* **55** 849
[24] Wilson R P 1969 *High Temp. Sci.* **1** 367
[25] Hoather W H 1936 *Proc. Phys. Soc.* **48** 706
[26] Ohsaka K, Chung S K, Rhim W K and Holzer J C 1997 *Appl. Phys. Lett.* **70** 423