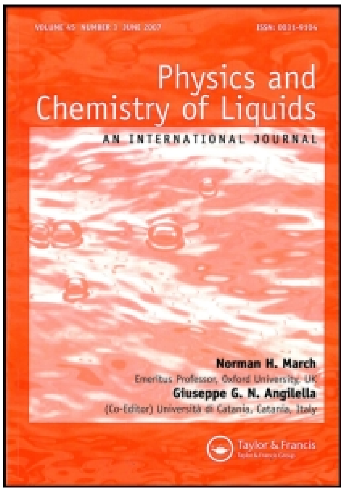

Physics and Chemistry of Liquids
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713646857

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To cite this Article Crawley, A. F.(1970) 'The viscosities of some lead-antimony and lead-antimony-tin alloys', Physics and Chemistry of Liquids, 2: 2, 77 — 85
To link to this Article: DOI: 10.1080/00319107008084081
URL: http://dx.doi.org/10.1080/00319107008084081

Physics and Chemistry of Liquids. 1970. Vol. 2, pp. 77–85

The Viscosities of Some Lead–Antimony and Lead–Antimony–Tin Alloys

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Received February 26, 1970

Abstract—The viscosities of some Pb–Sb alloys up to 10.8% Sb and Pb–Sb–Sn alloys up to 10% Sb and 10% Sn have been determined to an accuracy of ±0.5% by an absolute method applied to an oscillational viscosimeter. Andrade's equation $\eta v^{1/3} = A \exp C/vT$ has been used to describe the viscosity-temperature relationships, and values for the constants A and C are given. For Pb–Sb alloys, isothermal viscosities at 350 °C and 550 °C vary linearly with composition demonstrating ideal mixing. Using earlier data on Pb–Sn alloys, it is possible to demonstrate that the ternary alloys also show ideal solution behaviour over the composition range studied. Some brief comment is given on the state of reliable knowledge of alloy viscosities. It is concluded that much work remains to be done using tractable, accurate mathematical analyses for absolute viscosity measurements.

1. Introduction

This paper reports data for the viscosities of some alloys in the Pb–Sb and Pb–Sb–Sn systems. The work was undertaken as part of a long-term programme of liquid metals research at the Mines Branch, Department of Energy, Mines and Resources, Ottawa, Canada. The densities of Pb–Sb and Pb–Sb–Sn alloys have been reported in an earlier paper.(1)

Recent measurements from this laboratory(2) and by others(3) have presented overwhelming evidence of the ideal solution behaviour of Pb–Sn alloys.

2. Experimental Method

Viscosity coefficients were determined using an absolute technique

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applied to an oscillational viscosimeter. The mathematical analysis employed was that derived by Roscoe[4] for the case of a liquid contained in a closed right cylinder. Earlier papers[2,5] have described the apparatus, technique, and the procedures used to measure the experimental variables entering Roscoe's final working solution. During determinations of the logarithmic decrement, the viscosimeter was held under a vacuum of about 2×10^{-6} torr.

Measurements on the binary system were limited to hypoeutectic alloys ($<11.2\%$ Sb) because segregation effects, discussed in an earlier paper,[1] prevented the preparation of satisfactory ingots beyond the eutectic composition.

3. Results

Viscosities, measured over a range of temperatures up to 150 °C above the liquidus were plotted against temperature to yield the smooth curves evident in Figs. 1 and 2.

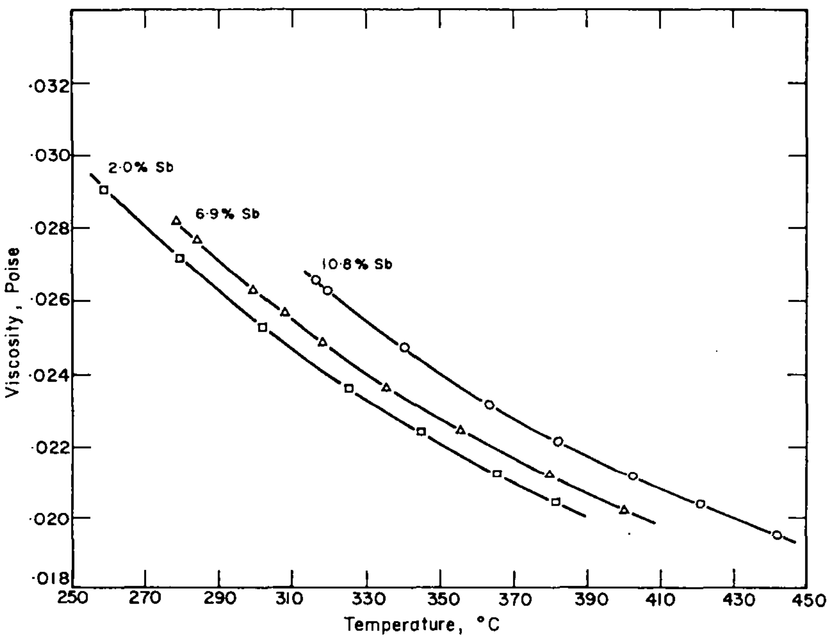

Figure 1. Viscosities of lead–antimony alloys as a function of temperature.

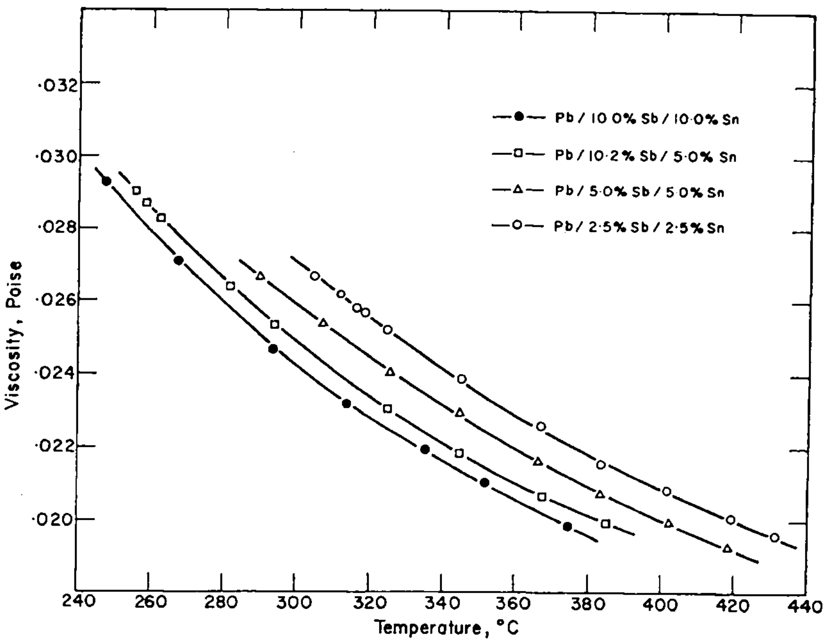

Figure 2. Viscosities of lead–antimony–tin alloys as a function of temperature.

A regression analysis was applied to the experimental results to give values for the constants A and C in the Andrade[6] equation.

$$\eta v^{1/3} = A \exp C/vT$$

where v is the specific volume and T is the absolute temperature. During the entire viscosity programme, Andrade's equation has been found to satisfactorily fit the data of the pure metals and alloys studied. Values of A and C for each alloy are given in Tables 1 and 2.

The results are accurate within $\pm 0.5\%$ considering known systematic errors and random errors based on deviations from Andrade's equation.

Table 1 Constants A and C in Andrade's Equation for Lead–Antimony Alloys

Wt. % Sb	$A \times 10^3$	C
2	2.428	89.561
6.9	2.368	91.030
10.8	2.373	91.969

Table 2 Constants A and C in Andrade's Equation for Lead–Antimony–Tin Alloys

Wt. % Sb	Wt. % Sn	$A \times 10^3$	C
2.5	2.5	2.502	88.255
5	5	2.469	89.728
10.2	5	2.242	96.327
10	10	2.192	98.848

4. Discussion

Earlier papers(2,5,7,8) from this laboratory on the viscosities of pure metals and alloys attest to the reliability of the data yielded by this

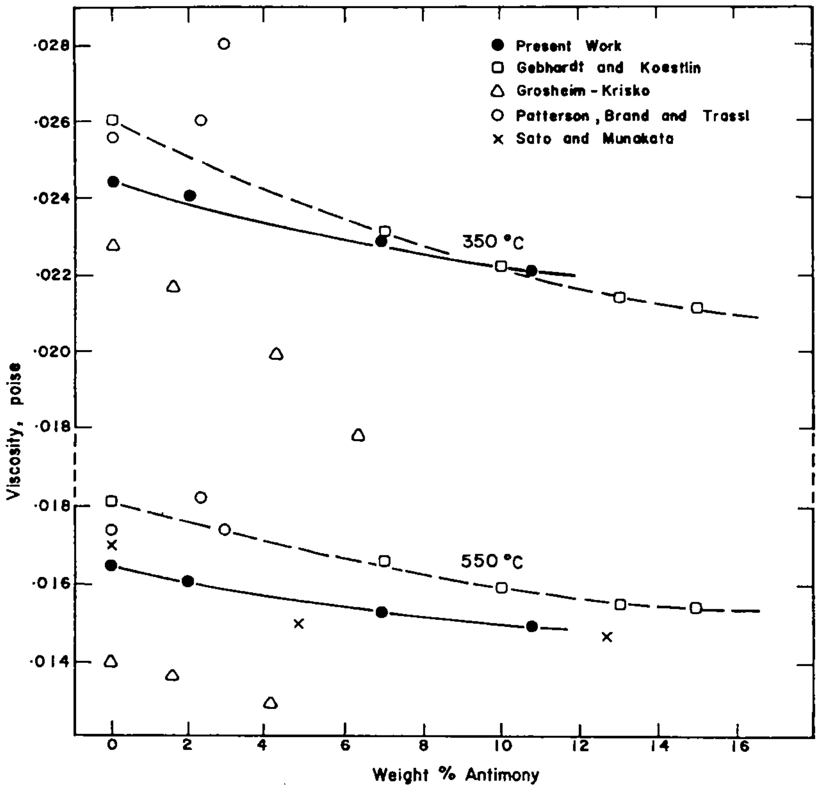

Figure 3. Viscosities of lead–antimony alloys at 350 °C and 550 °C against weight percentage of antimony.

technique. The present data, therefore, can be assumed to have equal reliability.

Lead–Antimony Alloys

The existing data[9,10,11,12] on this system are compared to the present results at 350 °C and 550 °C in Fig. 3. The evident lack of agreement among other workers probably arises from their use of calibrational techniques. Thresh[13] has fully discussed these methods and their inherent limitations in achieving absolute accuracy.

In Fig. 4, the isothermal viscosities determined at 350 °C and 550 °C have been plotted against molar composition. The evident linear relation-

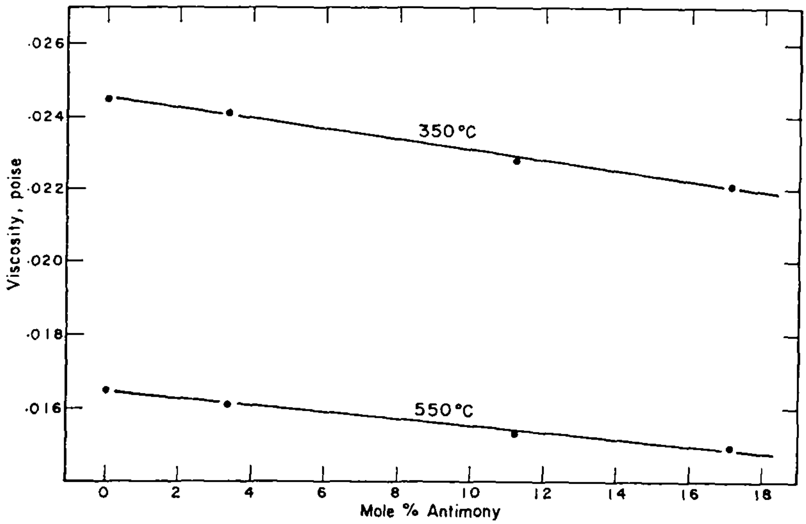

Figure 4. Viscosities of lead–antimony alloys at 350 °C and 550 °C against mole percentage of anitmony.

ship demonstrates the ideal solution behaviour of the system over this narrow composition range. These values, listed in column 3 of Table 3, were processed by the first-order regression analysis to yield the equations:

$$\eta = 0.02446 - 1.402\ 10^{-4}\ N_{Sb}\ (\text{at } 350\ °C)$$

$$\eta = 0.01644 - 9.174\ 10^{-5}\ N_{Sb}\ (\text{at } 550\ °C)$$

where N_{Sb} = mole % Sb. Values calculated from these equations are listed in column 4 of Table 3. The differences between these values and those calculated from Andrade's equation are shown in column 5 to be within the calculated limits of experimental error.

TABLE 3 Viscosities of Lead–Antimony Alloys at 350 °C and 550 °C Based on a Regression Analysis of Viscosities Calculated from Andrade's Equation

350 °C

Wt. % Sb	Mole % Sb	(1) Viscosity (η_1) (Andrade's Equation)	(2) Viscosity (η_2) (Regression Analysis)	$\frac{\eta_1 - \eta_2}{\eta_1} \times 100\%$
0	0	0.02441	0.02446	-0.20
2	3.356	0.02409	0.02399	0.42
6.9	11.200	0.02280	0.02289	-0.39
10.8	17.114	0.02210	0.02206	0.18

550 °C

Wt. % Sb	Mole % Sb	(1) Viscosity (η_1) (Andrade's Equation)	(2) Viscosity (η_2) (Regression Analysis)	$\frac{\eta_1 - \eta_2}{\eta_1} \times 100\%$
0	0	0.01651	0.01644	0.42
2	3.356	0.01610	0.01614	-0.24
6.9	11.200	0.01532	0.01542	-0.65
10.8	17.114	0.01494	0.01487	0.47

LEAD–ANTIMONY–TIN ALLOYS

A literature survey uncovered no data on this alloy system, thus no comparative assessment of the results is possible. However, some conclusions about the solution behaviour of these alloys can be drawn based on the ideal mixing shown by the binary lead–antimony and lead–tin[2] systems.

Assuming that the combined effects of tin and antimony on the viscosity of pure lead equals the sum of their individual effects, viscosities at 350 °C and 550 °C were calculated using the above equations for lead–antimony and those for the lead–tin system.[2] The values for pure lead were taken as the average at zero solute concentration given by both sets of equations. These data are compared with experimental values in Table 4. Differences compared with the measured values based on Andrade's equation are listed in the last column. Although in excess of 1% in some cases, these differences are not considered significant since the values have

TABLE 4 Viscosities of Lead–Antimony–Tin Alloys at 350 °C and 550 °C

350 °C

Wt. % Sb	Mole % Sb	Wt. % Sn	Mole % Sn	(1) Viscosity (η_1) (Andrade's Equation)	(2) Viscosity (η_2) (From Binary Data)	$\frac{\eta_1 - \eta_2}{\eta_1} \times 100\%$
2.5	4.106	2.5	4.212	0.02352	0.02348	0.17
5	7.935	5	8.139	0.02265	0.02252	0.57
10.2	15.654	5	7.871	0.02158	0.02147	0.51
10	14.866	10	15.249	0.02108	0.02080	1.33

550 °C

Wt. % Sb	Mole % Sb	Wt. % Sn	Mole % Sn	(1) Viscosity (η_1) (Andrade's Equation)	(2) Viscosity (η_2) (From Binary Data)	$\frac{\eta_1 - \eta_2}{\eta_1} \times 100\%$
2.5	4.106	2.5	4.212	0.01594	0.01582	0.75
5	7.935	5	8.139	0.01541	0.01523	1.17
10.2	15.654	5	7.871	0.01447	0.01453	−0.41
10	14.886	10	15.249	0.01412	0.01416	−0.28

been gathered by interpolation and extrapolation of three sets of experimental data. We may, therefore, conclude that, over the composition range studied, lead–antimony–tin alloys display simple ideal behaviour.

The work reported here together with recent investigations(2,3) represents a meaningful step towards some rationalisation of the viscosities of liquid alloys. A review on the structures of liquid metals by Wilson(14) points to the lack of good reliable alloy data. Much of the data in existence are not of recent origin and most have been determined by calibrational techniques. Evidently, much work on alloy systems remains to be done. The excellent mathematical analysis of Roscoe(4) applied to an oscillating closed right cylinder has been an important step.

Another interesting possibility has been the mathematical analysis of Armbruster[15] for the case of an open cylinder of liquid which, of course, introduces a meniscus factor. This method has yielded data for pure lead and tin[16] and for indium[17] very close to those determined in this laboratory.[2,8] Vignau, Azou and Bastien[18,19] have extended this mathematical treatment to yield a solution for the viscosity of a liquid covered by an immiscible layer of another liquid. They then used this analysis[20] to determine the viscosities of aluminium and aluminium–silicon alloys where the liquid metal was covered by a protective flux layer. The results for aluminium are the lowest recorded and the alloy data appear self-consistent. It would seem that these workers have succeeded in significantly reducing the oxidation problem which has hitherto impeded the gathering of definitive data on this metal.

Menz and Sauerwald[21] have used a double-capillary technique with considerable success to determine the viscosities of pure metals only. The method is truly absolute since it overcomes the need for kinetic energy and end corrections required by the single-capillary technique. Data reported for lead, tin and cadmium showed excellent agreement with results from this laboratory.[2,8]

Thus, at the present time there would appear to be at least three reliable absolute techniques for determining the viscosities of liquid metals and alloys. As the limitations of these techniques are different, the range of materials which can be investigated is quite wide. It will be interesting to see if the ideal solution behaviour found in lead–tin, lead–antimony and lead–antimony–tin alloys is operative in other systems, thus leading to a clearer understanding of the behaviour of liquid metals.

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