The Behavior of Mixing in Liquid Binary Alloys. Volume of Mixing in the Systems Lead-Tin, Lead-Antimony, and Lead-Thallium

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The molar volume of mixing in the liquid systems Pb-Sn, Pb-Sb, and Pb-Tl has been studied by means of silica glass pycnometer at 1045, 907, and 773 K respectively. The volumes of mixing in the systems Pb-Sn and Pb-Tl are slight positive (0.0263×10^{-6}—0.1767×10^{-6} m^3 mol^{-1}), while that in the system Pb-Sb is slight negative (-0.1524×10^{-6} m^3 mol^{-1}).

In a recent publication,[1] we disclosed some considerable points concerning the behavior of mixing in liquid binary alloys, i.e. dependence of the volume of mixing on the ratio of the atomic radii and electronegativity difference of the pure components. The existence of the compound Pb_3Bi_2 in the liquid Pb-Bi systems was deduced due to interpretations for the volume of mixing results and calculations using the Wagner's theory.[2] The present study describes further conclusion from the study on relative liquid binary lead alloys.

The relationship which has been derived by Predel and Emam[3] for the dependence of the volume of mixing on the atomic radii and electronegativity difference of the components in binary alloys is not reliable since such a relation gives unreasonable behavior of the volume of mixing. Also the theory by Prigogine[4] which gives the relation of the volume of mixing to other excess quantities, e.g. $\Delta_m H^\circ$ was derived for simple models since this theory assumes equality of the atomic radii of the components.

In view of the standpoints described above, a wide study on homologous series in the periodic table of elements is required in order to obtain systematic statements for the behavior of the volume of mixing. Thus we extend our previous study[1] on the system lead-bismuth to include the systems Pb-Sn, Pb-Sb, and Pb-Tl (see Ref. 5). These relative systems have been selected due to their completely miscibility in the liquid state (free from miscibility gaps).

Experimental

Sample. The metals used were of E. Merck extra purity grade. The preparation, treatment, and testing of the samples were all the same as described before.[1]

Apparatus. The apparatus used in the present study was a slightly modified version of the type used earlier,[1] which in turn was based on the construction used by Kleppa et al. (see Ref. 6). The 11—13 cm^3 silica glass container with a mass of 12—15 g and opening of 4 mm diameter is attached to a balance system by means of a high-temperature resisting wire of V2 A steel. The wire of V2 A steel goes through a tube of refined steel which in turn was fixed with the pycnometer. The role of the tube of V2 A steel was to immerse the pycnometer in the liquid sample existing inside a silica glass container. The lower end of the pycnometer is connected to a stirrer of refined steel being fixed in a plate of the same material. The role of this plate was to immerse the oxide layer which eventually exists at the surface area of the liquid to the bottom of the sample container. Both stirrer and plate were coated with a layer of aluminium oxide and its purpose was to prevent reaction with the sample. The pycnometer-sample assembly is suspended inside a double-walled V2 A steel shield system with enclosed heaters. The whole assembly except the cooling and balance systems is placed in a vertical tube furnace. The upper part of the shield is fixed in a ring of brass which in turn was fixed to the cooling system. Above the cooling system, there exists a balance system which was protected by means of a movable and evacuated plexiglass box. The apparatus is connected with argon gas supply which in turn was connected to a purification device. The role of the argon gas was to reduce the effect of oxidation at high temperatures.

The temperature differences between the corresponding parts of the calorimeter and also the temperature of the sample are measured by means of Pt-to-(Pt+10 mass per cent Rh) thermocouples. The calibration of the thermocouples was based on a standard Pt-PtRh thermocouple. The derived temperatures are judged to correspond with the IPTS-68 to within 0.01 K at 500 K and 0.1 K at 1000 K.

The calibration of the pycnometer was conducted at room temperature using sample of mercury (Suprapur, d=13.5377 g cm^{-3}, Merck Co.). The temperature was measured using a mercury-thermometer calibrated by the "physicalisch-Technischen Bundesanstalt, Germany". The balance used for calibration was a type of Mettler H 10 T.

Theory and Procedure. The additional behavior y^{ad} is given by

$$y^{ad}=y_1^\circ+x_2\,(y_2^\circ-y_1^\circ), \tag{1}$$

where y_1° and y_2° are the values of the molar quantities of the components 1 and 2, respectively. The form function of the excess function, X^y is given through the equation

$$X^y(x_2)=\frac{y^e}{x_2\,(1-x_2)}\cdot \tag{2}$$

y^e refers to the excess quantity.

The method used in the present study depends on the following procedure:

A silica glass pycnometer will be immersed in the liquid metallic sample inside a container filled with the protection gas. The pycnometer will be removed and immediately weighed.

The density d can be computed at the temperature of measurement ϑ from the mass m and the volume of the pycnometer V_{py} as follows:

$$d = m/V^{22.7} \cdot [1 + 3a(\vartheta - 22.7)], \qquad (3)$$

where 22.7 °C is the calibration temperature of the pycnometer and a the thermal coefficient of silica glass which is given by

$$a_{SiO_2} = 0.59 \times 10^{-6}\ \mathrm{K}^{-1}. \qquad (4)$$

Results and Discussion

In Table 1 are presented the results of the density determinations in the system Pb-Sn at 1040 K. The results have been least squares fitted by the following polynomial expression:

$$d(x_{Pb}) = 8.7463 + 0.4407x_{Pb} + 0.2045x_{Pb}^2 - 0.0677x_{Pb}^3. \qquad (5)$$

The standard deviation is 0.03% for a single measurement.

In Table 2 are listed the values of the average molar volume of the mixture Pb-Sn as determined in the present study. From curve-fitting calculations, the following polynomial function for the molar volume, $\bar{v}$, was derived.

$$\bar{v} = 13.5703 + 9.380x_{Pb} - 0.8142x_{Pb}^2 - 0.0079x_{Pb}^3 + 0.0131x_{Pb}^4 + 0.0234x_{Pb}^5. \qquad (6)$$

The estimated standard deviation is 0.011% for a single measurement.

Table 3 collects the values of the integral and partial molar volume of mixing as determined in the present study at 1040 K. The volume of mixing can be computed according to the following equation:

$$v^m = \bar{v} - v^{ad} = \bar{v} - v_1^\circ - x_2 \cdot (v_2^\circ - v_1^\circ), \qquad (7)$$

where v_1° and v_2° are the molar volumes of the pure components 1 and 2, respectively. From curve-fitting calculations, the following polynomial function is derived for the volume of mixing, v^m:

$$v^m = 0.0010 + 0.1368x_{Pb} - 0.03494x_{Pb}^2 + 0.5431x_{Pb}^3 - 0.4800x_{Pb}^4 + 0.1496x_{Pb}^5. \qquad (8)$$

Table 1. Experimentally Determined Densities (×10^{-3}) for Liquid Lead-Tin at 1040 K in kg m^{-3}. Estimated Standard Deviations in Parenthesis

x_{Pb}	d	x_{Pb}	d
0	8.7481(18)	0.6	9.0705(18)
0.1	8.7941(18)	0.7	9.1338(20)
0.2	8.8440(19)	0.8	9.1969(18)
0.3	8.9090(13)	0.9	9.2600(18)
0.4	8.9538(18)	1.0	9.3257(19)
0.5	9.0280(18)		

Table 2. Average Molar Volumes (×10^6) for the Mixture Pb-Sn in m^3 mol^{-1}. Estimated Standard Deviations in Parenthesis

x_{Pb}	$\bar{v}$	x_{Pb}	$\bar{v}$
0	13.5721(18)	0.6	18.9424(16)
0.1	14.5078(18)	0.7	19.7841(18)
0.2	15.4270(17)	0.8	20.2104(18)
0.3	16.3999(17)	0.9	21.4236(18)
0.4	17.2171(18)	1.0	22.2246(19)
0.5	18.0879(16)		

Table 3. Integral and Partial Molar Volume of Mixing (×10^6) for the Liquid Pb-Sn System at 1040 K in m^3 mol^{-1}. Estimated Standard Deviations in Parenthesis

x_{Pb}	v^m		v_{Sn}^m	v_{Pb}^m
	Present study	Crawley[10]		
0[a]	0	0.0029	0.0010	0.1378
0.1	0.0134(17)	0.0158	0.0035	0.0849
0.2	0.0197(17)	0.0230	0.0084	0.0565
0.3	0.0233(16)	0.0251	0.0133	0.0414
0.4	0.0256(18)	0.0279	0.0181	0.0324
0.5	0.0263(17)	0.0275	0.0239	0.0254
0.6	0.0258(18)	0.0283	0.0326	0.0185
0.7	0.0234(17)	0.0266	0.0448	0.0118
0.8	0.0189(16)	0.0219	0.0622	0.0061
0.9	0.0121(17)	0.0142	0.0836	0.0023
1.0[a]	0	0.0033	0.1058	0.0011

a) Extrapolated value.

According to the definition of the partial molar quantities (see Ref. 1), the following polynomial functions are derived for the partial molar volumes of mixing of tin and lead:

$$v_{Sn}^m = 0.0010 + 0.3494x_{Pb}^2 - 1.086x_{Pb}^3 + 1.4400x_{Pb}^4 - 0.5984x_{Pb}^5, \qquad (9)$$

$$v_{Pb}^m = 0.1378 - 0.6988x_{Pb} + 1.9787x_{Pb}^2 - 3.0062x_{Pb}^3 + 2.1880x_{Pb}^4 - 0.5984x_{Pb}^5. \qquad (10)$$

From Table 3, we conclude that the estimated standard devitation (e.s.d.) does not exceed that expected

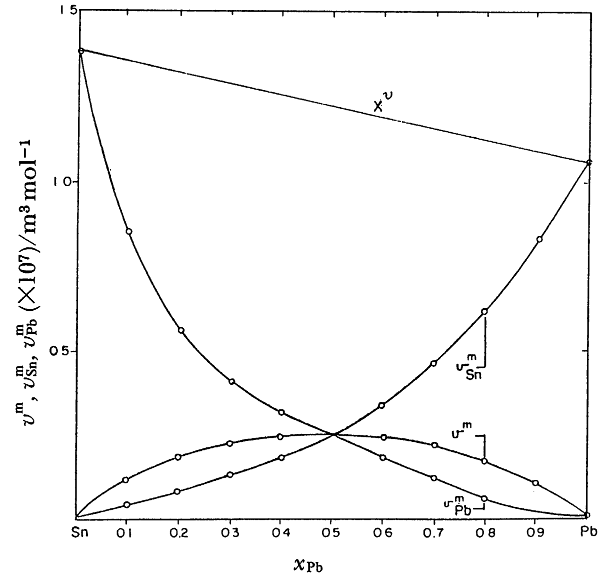

Fig. 1. Integral and partial molar volume of mixing for the liquid Pb-Sn system at 1040 K.

from the pycnometer measurement uncertainties, 2.2×10^{-9} to $2.4\times10^{-9}\,\mathrm{m^3\,mol^{-1}}$. Figure 1 shows the curves of the volume of mixing, v^{m} against the mole fraction of lead, x_{Pb}. It is clearly observable that the volume of mixing takes slight positive values which appear to be smaller than those found for the related system Bi–Pb.[1)]

The volume of mixing in the liquid system Pb–Sn shows a positive trend with a maximum value at about $x_{\mathrm{Pb}}=0.5$ which appears to be similar to that of the enthalpy of mixing.[7,8)]

The results obtained by Predel and Emam[9)] for the volume of mixing in the system Pb–Sn and those by Crawley[10)] showed a considerable discrepancy between them. It appears that the measurement technique used by these authors could not overcome the oxidation problem. This would be clear from the construction of the apparatus and the method described by them. The effect of oxidation led to a considerable percentage error in the results. However, the composition of the maximum of the volume of mixing as determined in both studies is compatible to that found in the present study. The results for v^{m} obtained by Crawley[10)] seemed to be more close to those obtained in the present study rather than those by Predel and Emam.[9)] Values for v^{m} obtained by Crawley[10)] are given in Table 3.

Table 4 represents values for the form function of the volume of mixing at 1040 K which is given by the following linear function:

$$X^{v}=0.1368-0.1496x_{\mathrm{Pb}}. \qquad (11)$$

Table 4. The Form Function of the Volume of Mixing ($\times10^6$) for the Liquid Pb-Sn System at 1040 K in $\mathrm{m^3\,mol^{-1}}$. Estimated Standard Deviations in Parenthesis

x_{Pb}	X^{v}	x_{Pb}	X^{v}
0	0.1368(15)	0.6	0.0470(15)
0.1	0.1218(16)	0.7	0.032(14)
0.2	0.1069(16)	0.8	0.0171(16)
0.3	0.0919(14)	0.9	−0.0221(13)
0.4	0.0770(17)	1.0	−0.0128(18)
0.5	0.0620(15)		

Table 5. Experimentally Determined Densities ($\times10^{-3}$) for Liquid Lead-Antimony at 907 K in $\mathrm{kg\,m^{-3}}$. Estimated Standard Deviations in Parenthesis

x_{Sb}	d	x_{Sb}	d
0	8.5306(17)	0.6	8.3306(16)
0.1	8.4896(17)	0.7	8.3062(17)
0.2	8.4526(16)	0.8	8.2820(16)
0.3	8.4190(18)	0.9	8.2588(17)
0.4	8.3877(17)	1.0	
0.5	8.3575(17)		

Table 6. Average Molar Volume ($\times10^6$) for the Mixture Pb-Sb at 907 K in $\mathrm{m^3\,mol^{-1}}$. Estimated Standard Deviations in Parenthesis

x_{Sb}	$\bar{v}$	x_{Sb}	$\bar{v}$
0	24.2933(23)	0.6	18.7207(22)
0.1	23.4040(23)	0.7	17.8200(23)
0.2	22.5930(22)	0.8	16.9344(22)
0.3	21.7072(22)	0.9	15.8804(24)
0.4	20.7229(22)	1.0	14.2146(22)
0.5	19.7024(23)		

Table 7. Integral and Partial Molar Volume of Mixing ($\times10^6$) for the Liquid Pb-Sb System at 907 K in $\mathrm{m^3\,mol^{-1}}$. Estimated Standard Deviations in Parenthesis

x_{Sb}	v^{m}		$v^{\mathrm{m}}_{\mathrm{Pb}}$	$v^{\mathrm{m}}_{\mathrm{Sb}}$
	Present study	O. Sato[12)]		
0[a)]	0	−0.0051	−0.0017	−0.0705
0.1	−0.0150(24)	−0.0190	+0.0050	−0.1954
0.2	−0.0437(24)	−0.0482	+0.0224	−0.2960
0.3	−0.0789(24)	−0.0825	+0.0384	−0.3371
0.4	−0.1151(25)	−0.1194	+0.0209	−0.3128
0.5	−0.1424(24)	−0.1463	−0.0401	−0.2400
0.6	−0.1524(24)	−0.1565	−0.1556	−0.1464
0.7	−0.1394(24)	−0.1440	−0.3122	−0.0621
0.8	−0.1028(23)	−0.1071	−0.4655	−0.0101
0.9	−0.0526(24)	−0.0566	−0.5309	+0.0032
1.0[a)]	0	−0.0039	−0.3738	−0.0024

a) Extrapolated value.

Table 5 summarizes values for the density of the system Pb–Sb at 907 K. The results have been least squares fitted by the following polynomial function:

$$d(x_{\mathrm{Sb}})=8.5299-0.4298x_{\mathrm{Sb}}+0.2003x_{\mathrm{Sb}}^{2}-0.066x_{\mathrm{Sb}}^{3}. \qquad (12)$$

The standard deviation is 0.02% for a single measurement.

Table 6 gives results for average molar volume of the mixture Pb–Sb at 907 K. From curve-fitting calculation, the following polynomial function of $\bar{v}$ was derived:

$$\bar{v}=24.2910-10.2534x_{\mathrm{Sb}}+20.2464x_{\mathrm{Sb}}^{2}-76.3795x_{\mathrm{Sb}}^{3}+104.3846x_{\mathrm{Sb}}^{4}-48.0767x_{\mathrm{Sb}}^{5}. \qquad (13)$$

The estimated standard deviation is 0.013% for a single measurement.

Results of the determinations on the volume of mixing at 907 K in the system Pb–Sb are presented in Table 7 together with those for the partial molar volume of mixing of Pb and Sb. The following polynomial functions were derived for v^{m}, $v^{\mathrm{m}}_{\mathrm{Pb}}$, and $v^{\mathrm{m}}_{\mathrm{Sb}}$ to produce the curves as shown in Fig. 2:

$$v^{\mathrm{m}}=-0.0017-0.0688x_{\mathrm{Sb}}-0.5773x_{\mathrm{Sb}}^{2}-0.9854x_{\mathrm{Sb}}^{3}+3.6030x_{\mathrm{Sb}}^{4}-1.9722x_{\mathrm{Sb}}^{5}, \qquad (14)$$

$$v^{\mathrm{m}}_{\mathrm{Pb}}=-0.0017+0.5773x_{\mathrm{Sb}}^{2}+1.9708x_{\mathrm{Sb}}^{3}-10.8090x_{\mathrm{Sb}}^{4}+7.8888x_{\mathrm{Sb}}^{5}, \qquad (15)$$

$$v^{\mathrm{m}}_{\mathrm{Sb}}=-0.0705-1.1546x_{\mathrm{Sb}}-2.3789x_{\mathrm{Sb}}^{2}+16.3828x_{\mathrm{Sb}}^{3}-20.6700x_{\mathrm{Sb}}^{4}+7.8888x_{\mathrm{Sb}}^{5}. \qquad (16)$$

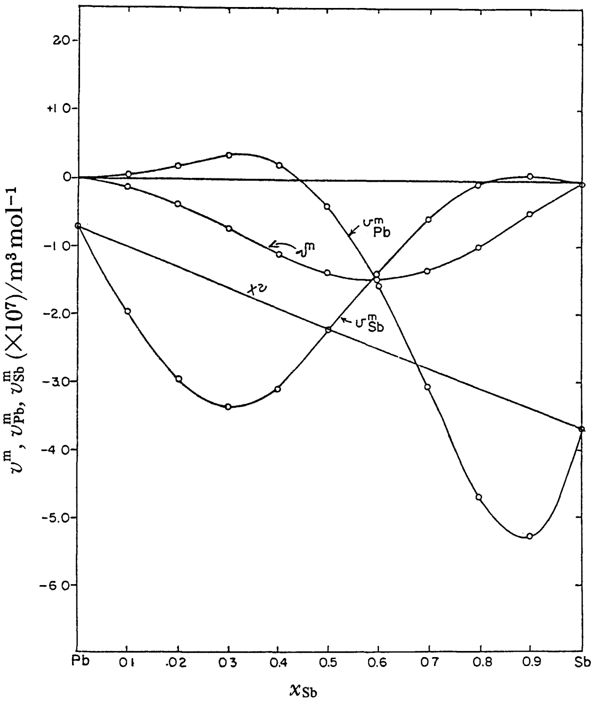

Fig. 2. Integral and partial molar volume of mixing for the liquid Pb-Sb system at 907 K.

Table 7 shows that the e.s.d. is less than expected from the pycnometer measurement uncertainties as given above. Table 7 also indicates the slightly negative volume of mixing in the system Pb-Sb.

The volume of mixing and its partial molar quantities in the liquid system Pb-Sb show trends which are similar to those of the enthalpy of mixing.[8,11] Both v^m and $\Delta_m H^\circ$ are slight negative and show a minimum at almost same composition.

A slightly negative volume of mixing for Pb-Sb bas also been observed by Sato.[12] The result obtained by this author is in acceptable agreement with the present one (see Table 7). Also the value obtained by Crawley[10] is in fair agreement with the present one.

Table 8 demonstrates the values obtained for the form functions of the volume of mixing in the system Pb-Sb at 907 K. The form function, X^v is expressed by the following linear polynomial function:

$$X^v = -0.0688 - 0.3050\,x_{Sb}. \tag{17}$$

Table 8. The Form Function of the Volume of Mixing (×10⁶) for the Liquid Pb-Sb at 907 K in m³ mol⁻¹. Estimated Standard Deviations in Parenthesis

x_{Sb}	$-X^v$	x_{Sb}	$-X^v$
0	0.0688(0)	0.6	0.2178(20)
0.1	0.0973(20)	0.7	0.2842(19)
0.2	0.1277(21)	0.8	0.3147(19)
0.3	0.1623(20)	0.9	0.3453(20)
0.4	0.1928(20)	1.0	0.3737(1)
0.5	0.2232(168)		

Table 9. Experimentally Determined Densities (×10⁻³) for Liquid Lead-Thalium at 775 K in kg m⁻³. Estimated Standard Deviations in Parenthesis

x_{Pb}	d	x_{Pb}	d
0	10.9763(20)	0.6	10.7189(20)
0.1	10.9230(15)	0.7	10.6844(21)
0.2	10.8752(19)	0.8	10.6553(20)
0.3	10.8303(20)	0.9	10.6249(15)
0.4	10.7909(20)	1.0	10.5960(20)
0.5	10.7537(21)		

Table 10. Average Molar Volume (×10⁶) for the Mixture Pb-Tl at 775 K in m³ mol⁻¹. Estimated Standard Deviations in Parenthesis

x_{Pb}	$\overline{v}$	x_{Pb}	$\overline{v}$
0	18.6244(18)	0.6	19.2301(18)
0.1	18.7579(00)	0.7	19.3141(17)
0.2	18.8490(20)	0.8	19.3981(19)
0.3	18.9519(18)	0.9	19.4804(20)
0.4	18.0494(19)	1.0	19.5600(18)
0.5	19.1420(20)		

Table 11. Integral and Partial Molar Volume of Mixing (×10⁶) for the Liquid Pb-Tl System at 775 K in m³ mol⁻¹. Estimated Standard Deviations in Parenthesis

x_{Pb}	v^m		v^m_{Tl}	v^m_{Pb}
	Present study	Predel & Emam[9]		
0[a]	0	0.0022	0.0010	−0.0069
0.1	0.0090(16)	0.0117	−0.0053	+0.1214
0.2	0.0267(17)	0.0292	−0.0192	+0.2019
0.3	0.0528(20)	0.0560	−0.0367	+0.2546
0.4	0.0842(17)	0.0571	−0.0529	+0.2856
0.5	0.1190(17)	0.1222	−0.0570	+0.2915
0.6	0.1516(16)	0.1553	−0.0255	+0.2670
0.7	0.1767(16)	0.1794	+0.0835	+0.2101
0.75	0.1767(16)	0.1801	+1.1867	+0.1712
0.8	0.1647(17)	0.1682	+0.3363	+0.1281
0.9	0.1240(16)	0.1271	+0.8294	+0.0439
1.0[a]	0	0.0039	+1.6961	+0.0016

a) Extrapolated value.

Results of the density determination in the system Pb-Tl at 775 K are presented in Table 9. The results have been least squares fitted by the following polynomial function:

$$d(x_{Sb}) = 10.9743 - 0.5530x_{Sb} + 0.2577x_{Sb}^2 - 0.0850x_{Sb}^3. \tag{18}$$

The standard deviation is 0.04% for a single measurement.

Table 10 collects values of the average molar volume of mixture Pb-Tl at 775 K as determined in the present study. Using curve fitting calculation, the following polynomial function for, $\overline{v}$, was derived:

$$\overline{v} = 18.6226 + 1.1831x_{Pb} - 0.2165x_{Pb}^2 + 0.0812x_{Pb}^3 + 0.1927x_{Pb}^4 - 0.0625x_{Pb}^5. \tag{19}$$

The estimated standard deviation is 0.014% for a single

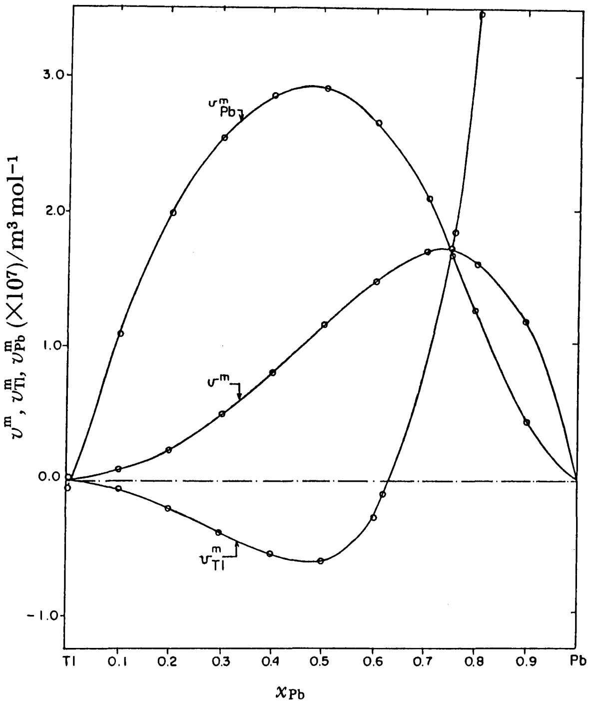

Fig. 3. Integral and partial molar volume of mixing for the liquid Pb-Tl system at 775 K.

measurement.

Values of the integral and partial molar volumes of mixing in the system Pb-Tl at 775 K are listed in Table 11. Appropriate polynomial functions were derived to produce the curves of v^m, v^m_{Tl}, and v^m_{Pb} as illustrated in Fig. 3:

$$v^m = 0.0010 - 0.0079x_{Pb} + 0.8185x^2_{Pb} + 1.1643x^3_{Pb} - 1.2479x^5_{Pb}, \quad (20)$$

$$v^m_{Tl} = 0.0010 - 0.8185x^2_{Pb} + 2.3286x^3_{Pb} - 4.8066x^4_{Pb} + 4.9916x^5_{Pb}, \quad (21)$$

$$v^m_{Pb} = -0.0069 + 1.6370x_{Pb} - 4.3114x^2_{Pb} + 8.7374x^3_{Pb} - 11.0461x^4_{Pb} + 4.9916x^5_{Pb}. \quad (22)$$

The estimated standard deviations in parenthesis, as given in Table 11, show that the e.s.d. does not exceed that from pycnometer measurement uncertainties, 2.2X10^{-9} to 2.4X10^{-9} m^3 mol^{-1}.

The v^m is slight positive while $\Delta_m H^\circ$ is slight negative. The maximum of the volume of mixing is situated at compostion $x_{Pb} = 0.75$ and the minimum of enthalpy of mixing is at x_{Pb}=0.5.

Table 12. The Form Function of the Volume of Mixing (X10^6) for the Liquid Pb-Tl at 775 K in m^3 mol^{-1}. Estimated Standard Deviations in Parenthesis

x_{Pb}	X^v	x_{Pb}	X^v
0	−0.099(20)	0.6	+0.7430(22)
0.1	+0.1184(25)	0.7	+0.8676(20)
0.2	+0.2442(25)	0.75	+0.9301(21)
0.3	+0.3689(24)	0.8	+0.9926(22)
0.4	+0.4933(20)	0.9	+1.1172(20)
0.5	+0.6161(00)	1.0	+1.1709(20)

The present value of the volume of mixing is in reasonable agreement with that reported by Predel and Emam.[9] Their interpretetion of the volume of mixing in the system Tl-Pb is compatable to that of the present study.

Table 12 presents values of the form function of the volume of mixing in the system Pb-Tl at 775 K. The values of X^v have been obtained from calculations using the following linear function:

$$X^v = -0.0079 + 1.2479x_{Pb}. \quad (23)$$

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