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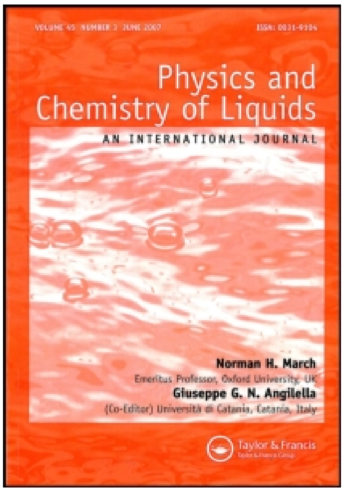

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Surface Tension and Sound Velocity of Pb–Sn Alloys in the Liquid State

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An attempt has been made to evaluate the surface tension and sound velocity of Pb–Sn alloys in the liquid state over a wide range of temperature and composition in the light of Flory's statistical theory. The agreement between theoretical and experimental values is quite satisfactory.

Successful attempts have been made by several investigators to calculate theoretically the surface tension of liquids and liquid mixtures. A critical review is given by Ono *et al.*[1] and Gubbins *et al.*[2] Amongst the various theories utilized for this purpose, Flory theory[3–5] has been found to be the most adequate. Many investigators[6–11] used the Flory theory for the theoretical computation of surface tension of liquids and liquid mixtures. Theoretical evaluation of sound velocities in liquids and liquid mixtures from Flory theory[3–5] in conjunction with Auerbach's relation[12] has been discussed by Pandey[13] and Mishra.[14] Pandey *et al.*[15–18] have applied the Flory statistical theory in the case of molten salts and their binary mixtures for the evaluation of surface tension, sound velocity and some other thermodynamic properties. Recently Pandey *et al.*[19] and Chaturvedi *et al.*[20] have successfully applied this theory in case of liquid metal and liquid mercury at elevated pressures. It is clear from the literature that no attempt has been so far, been made to evaluate the surface tension and sound velocity for metal alloys using Flory's[3–5] theory. Although Konyuchenko[21] have evaluated theoretically the surface tension and sound velocity of Pb–Sn alloys, but they did not utilize the Flory theory for this purpose. The present paper deals with the application of Flory theory for the theoretical evaluation of surface tension and sound velocity of Pb–Sn alloys.

THEORETICAL

According to Auerbach[12] relation, the sound velocity, U is expressed by the equation

$$U = \left(\frac{\sigma}{6.3 \times 10^{-4}\,\rho}\right)^{2/3} \tag{1}$$

where σ and ρ are the surface tension and density respectively. According to Flory theory the surface tension, σ, is expressed as

$$\sigma = \sigma^{*}\tilde{\sigma}(\tilde{V}) \tag{2}$$

where σ^{*} and $\tilde{\sigma}(\tilde{V})$ are the characteristic surface tension and reduced surface tension respectively. The characteristic surface tension, as given by Patterson and Rastogi[6] in their extension of corresponding state theory,[22,23] can be expressed as

$$\sigma^{*} = k^{1/3}p^{*2/3}T^{*1/3} \tag{3}$$

Here k is the Boltzmann constant, p^{*} the characteristic pressure and T^{*} the characteristic temperature. Starting with the work of Prigogine and Sarga,[22] using Flory's statistical model of liquid state, the equation for reduced surface tension[6] can be given as

$$\tilde{\sigma}(\tilde{V}) = M\tilde{V}^{-5/3} - \frac{\tilde{V}^{1/3} - 1}{\tilde{V}^{2}} \ln \frac{\tilde{V}^{1/3} - 0.5}{(\tilde{V}^{1/3} - 1)} \tag{4}$$

where $\tilde{V}$ is the reduce volume and M is the fractional decrease in the neighbourhood of a cell in the surface phase as compared to the bulk phase, its most suitable value is 0.29. Pandey[13] and Mishra[14] have suggested some modification to its previous values in order to obtain better results at elevated pressures. Recently Pandey *et al.*[15,19] and Chaturvedi *et al.*[20] used an alternative value in the case of molten salts and liquid metals. This value differs from those used for molecular liquids and liquid mixtures. In the present paper we have used 0.42 for M. Equations (3) and (4) have been derived in the light of the reduced equation of state[3–5]

$$\tilde{V} = \left[\frac{\alpha T}{(3 + 3\alpha T)} + 1\right]^{3} \tag{5}$$

$$P^{*} = \frac{\alpha T \tilde{V}^{2}}{\beta_T} \tag{6}$$

$$T^{*} = \frac{T\tilde{V}^{4/3}}{\tilde{V}^{1/3} - 1} \tag{7}$$

where α and β_T are the thermal expansion coefficient and isothermal compressibility respectively.

For binary system

$$\tilde{V} = V_{\mathrm{mix}}/\sum X_i V_i^* \tag{8}$$

where X_i is the mole fraction, V_i^* is the characteristic volume of the pure component 1, V_{mix} is the molar volume of the mixture and $V_i^* = V_i/\tilde{V}_i$.

$$P^* = \psi_1 P_1^* + \psi_2 P_2^* - \psi_1 \theta_2 X_{12} \tag{9}$$

where ψ_1 and ψ_2 are the segment fractions. θ_2 is the site fraction of component 2; X_{12} is the interaction parameter. P_1^* and P_2^* are the characteristic pressures of pure components comprising the mixture. The segment fraction as per Flory theory[3,4] has been evaluated from the relation.

$$\psi_2 = 1 - \psi_1 = \frac{X_2}{X_2 + X_1 V_1^*/V_2^*} \tag{10}$$

where X_1 and X_2 are the atomic concentrations of the component and V_1^* and V_2^* are the characteristic volume of the pure components. The site fraction is calculated from the relation.

$$\theta_2 = 1 - \theta_1 = \frac{\psi_2}{\psi_2 + \psi_1 (V_2^*/V_1^*)^{1/3}} \tag{11}$$

The interaction parameter X_{12} has been evaluated from the Berthelots' approximation for monopolar species and is expressed as

$$X_{12} = P_1^*\left[1 - \left(\frac{V_2^*}{V_1^*}\right)^{1/6}\left(\frac{P_2^*}{P_1^*}\right)^{1/3}\right]^2 \tag{12}$$

RESULTS AND DISCUSSION

The values of reduced volume, $\tilde{V}$ and characteristic pressure, P^*, as obtained through Eqs (8) and (9) respectively for Pb–Sn alloys in liquid state over a wide range of temperature and compositions, are presented in Table I. The essential data required for the calculation of these values have been taken from the literature.[21] The values of reduced surface tension and characteristic surface tension have been obtained with the help of these parameters, viz., $\tilde{V}$, P^* and T^* vide Eqs (8), (9) and (7) respectively. These values of σ^* and $\tilde{\sigma}(\tilde{V})$ have been utilized to obtain the values of surface tension, of Pb–Sn alloys, which are presented in Table I with their experimental values.[24,25] Both the experimental and theoretical values of surface tension are represented graphically in Figure 1 as a function of molefraction

TABLE I
Reduced volume, characteristic pressure, calculated and experimental values of surface tension and sound velocity of Pb–Sn alloy in liquid state

T/K	X_2	$\tilde{V}$	P^* (108)	σ (Cal.) dyne/cm	σ (exp.) dyne/cm	U Cal. m/sec.	U (exp.) m/sec.
673	0.1	1.0520	275.77	516.0	519	2369.6	2360
	0.2	1.0764	271.66	467.7	501	2168.2	2315
	0.3	1.0983	267.66	429.7	482	2002.8	2235
	0.45	1.1086	261.88	408.2	475	1925.9	2175
	0.60	1.1199	256.34	387.3	460	1715.8	2080
873	0.1	1.0729	349.11	566.9	507	2549.5	2320
	0.2	1.0926	345.55	525.1	497	2373.1	2270
	0.3	1.1195	342.08	477.5	478	2169.6	2190
	0.45	1.1264	337.04	459.7	467	2045.1	2120
	0.60	1.1485	332.19	424.6	451	1855.8	2030
973	0.1	1.0830	386.13	589.2	502	2641.2	2305
	0.2	1.1067	381.08	539.1	495	2430.8	2250
	0.3	1.1286	376.15	498.0	476	2261.5	2170
	0.45	1.1474	369.01	462.9	463	2070.3	2095
	0.60	1.1599	362.15	438.5	447	1908.2	2005
Average % error				7.2		7.4	

* The alloy compositions are given in wt %

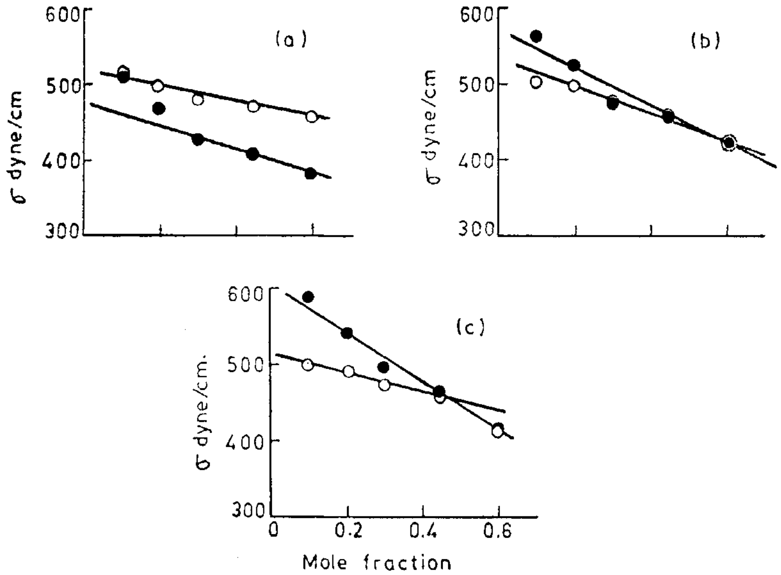

FIGURE 1 Theoretical (●) and experimental (○) values of the surface tension as a function of mole fraction X_2 for various temperature (a) 673°K, (b) 873°K, (c) 973°K.

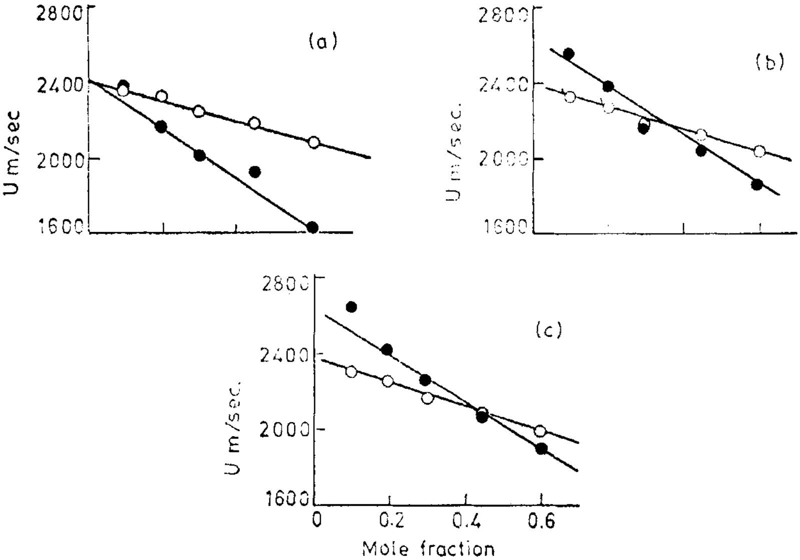

FIGURE 2 Theoretical (●) and experimental values (○) of the velocity as a function of mole fraction X_2 for various temperature (a) 673°K, (b) 873°K, (c) 973°K.

at all three temperatures. An inspection of the columns fifth and sixth of Table I and of Figure 1 reveals that the average percentage deviation between the theoretical and experimental values is not more than 7.2. The values of sound velocity, U, as obtained from Eq. (1) are given in the seventh column of Table I with their experimental values.[21] Although Eq. (1) is empirical in nature, its validity is well justified,[26] as it gives a maximum error of only $\pm 4\%$ for theoretical sound velocities if all the experimental data are precise. Theoretical and experimental values of sound velocity are also presented graphically in Figure 2 as a function of molefraction at different temperature. The average percentage deviation between theoretical and experimental values of sound velocity is not more than 7.5. It is clear from the Table I, Figures 1 and 2 that the theoretical values of surface tension and sound velocity of Pb–Sn alloys in liquid state obtained from Flory's statistical theory, give quite satisfactory agreement with the experimental and other theoretical[21] values. The results of the present calculation show that the rate of surface tension and sound velocity with temperature is positive, whereas experimentally it is negative. Our main aim is to show the general applicability of the Flory theory in a qualitative way. It appears that Flory theory cannot explain the reverse trend of sound velocity and surface tension with temperature, although it is clear from the table and graphs 1 and 2 that at different molefraction, variation of surface tension and sound velocity predicted theoretically is similar to those obtained from experimentally. We can also arrive at the conclusive juncture that Flory's theory has universal applicability and it can also be applied in the case of liquid metal alloys over a wide range of temperatures at different molefractions.

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