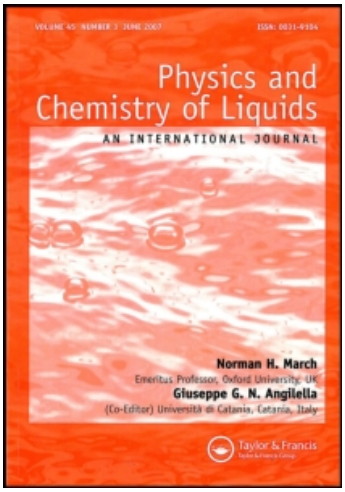

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Sound velocity, effective Debye temperature and pseudo-Grüneisen parameters of Pb-Sn mixtures at elevated temperatures

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Sound velocity, effective Debye temperature and pseudo-Grüneisen parameters of Pb–Sn mixtures at elevated temperatures

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Sound velocity and allied parameters, effective Debye temperature and pseudo-Grüneisen parameter for Pb–Sn molten binary mixture have been studied theoretically over a wide range of temperature and composition. An interaction study has also been made in the present context by computing excess pseudo-Grüneisen parameter which is a measure of the extent of molecular interaction involved in the liquid mixture. Effect of temperature on intermolecular free length has been studied with the help of free length theory of Jacobson.

Keywords: Molten binary mixture; Grüneisen parameters; Sound velocity

1. Introduction

Successful attempts have been made by a number of workers [1–12] on the measurements and theoretical prediction of sound velocity, effective Debye temperature and pseudo-Grüneisen parameter in liquid mixtures using various liquid models. However, there are only a few measurements of sound velocity of liquid binary mixtures [13]. The study of atomic motions in liquids plays a very significant role in understanding the solid-like behaviour of liquids. But unfortunately, no successful theory is yet available to explain such complicated behaviour of liquids. The dynamical behaviour of liquids can be studied by the accurate measurements of the energy changes due to the scattering. This technique was further used by Hughee *et al*. [9] and Frenkel [8]. Joshi [6] and Singwi and Sjalander [7] evaluated the Debye temperature of water at a particular temperature, in good agreement with the values given by Singwi *et al*. [7]. Such types of calculation have recently been extended to other liquids [5] assuming the atomic motions in these liquids are similar to those of solids.

In the theory of lattice dynamics, a diagnostic parameter found to be very useful in the study of the thermodynamics and related properties of crystalline lattices was the

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Grüneisen parameter [14]. Knopoff and Shapiro [10], using its pseudo counterpart, extended its use in the study of liquids. Since then, it has become an important tool in estimating the internal structures, clustering phenomenon and other quasi-crystalline properties of liquids [15–16].

In the present investigation, sound velocity and allied parameters have been predicted theoretically for a Pb–Sn molten binary mixture with the help of the collision factor theory due to Schaaff [17] and the free length theory due to Jacobson [18] at elevated temperatures over a wide range of composition. Successful attempts using Flory theory [19,20] have been employed to evaluate the pseudo-Grüneisen parameter of the pure metals together with the binary molten mixture and normal alkane quaternary liquid mixture for estimating the structural changes during mixing. With the use of ultrasonic absorption data, effective Debye temperature of the mixture Pb–Sn have been evaluated as a function of temperature assuming the quasi-crystalline model for liquids over a wide range of composition. An interaction study has also been made in the present context. The validity of these theories lies on the fact that there is a good agreement between experimental and theoretical values of sound velocity.

2. Theoretical

Schaaff developed the following expression for evaluating the sound velocity in pure liquids which in turn has been extended by Nutseh Kuhnkies [21] for binary liquid mixtures

$$U_{\mathrm{m}} = U_{\infty} S_{\mathrm{m}} r_{f_{\mathrm{m}}} \tag{1}$$

where $U_{\infty} = 1600\,\mathrm{m\,s^{-1}}$, S_{m} is the collision factor and $r_{f_{\mathrm{m}}}$ is the space filling factor of the liquid mixture respectively. The latter can be obtained by the relation

$$r_{f_{\mathrm{m}}} = \frac{B_{\mathrm{m}}}{V_{\mathrm{m}}} \tag{2}$$

where B_{m} is the actual volume/mole and V_{m} is the molar volume of the liquid mixture.

The actual volume of the liquid mixture is given by the expression

$$B_{\mathrm{m}} = \frac{3}{4}\pi r_{\mathrm{m}}^{3} N \tag{3}$$

where r_{m} is molecular radius and N is Avogadro's number. The volume associated with the molecular radius can be obtained from the formula given by Schaaff and Gopala Rao [22].

According to Eyring *et al.* [23], the intermolecular free length is given by the relation

$$L_{\mathrm{f}} = \frac{2V_{\mathrm{a}}}{y} \tag{4}$$

where V_{a} is the available molar volume, equal to $V_{\mathrm{a}} = V_{\mathrm{t}} - V_{\mathrm{o}}$. Here V_{t} and V_{o} denote the molar volumes at absolute temperature and at absolute zero respectively, at a particular pressure while y is the surface area mole^{-1}. These quantities V_0 and y can be

expressed as follows,

$$V_0 = V_T\left(1 - \frac{T}{T_c}\right)^{0.3} \quad \text{and} \quad y = (36\pi N V_0^2)^{1/3} \tag{5}$$

The effective Debye temperature θ can be evaluated by using the following expression [24]

$$\theta = \frac{h}{k}\left[\frac{9N/4\pi V}{(1/U_\ell^3) + (2/U_t^3)}\right]^{1/3} \tag{6}$$

where U_ℓ and U_t are the propagation velocities for longitudinal and transverse modes respectively. V, the molar volume and h, k and N are respectively the Planck's constant, Boltzmann's constant and Avogadro's number.

The two wave velocities may be expressed in terms of density (ρ), the instantaneous adiabatic compressibility ($\beta_{a,\infty}$) and Poisson's ratio (σ), for liquids exhibiting the quasi-crystalline properties, as follows;

$$\frac{1}{U_\ell^3} + \frac{2}{U_t^3} = (\beta_{a,\infty}\rho)^{3/2}\left[\left\{\frac{1+\sigma}{3(1-\sigma)}\right\}^{3/2} + 2\left\{\frac{2(1+\sigma)}{3(1-2\sigma)}\right\}^{3/2}\right] \tag{7}$$

and

$$\beta_{a,\infty} = \left[\beta_{T,\infty} - \frac{T\alpha^2 V}{C_p}\right] \tag{8}$$

where α is the coefficient of linear expression, $\beta_{T,\infty}$, the isothermal compressibility and C_p the specific heat at constant pressure. On neglecting the second term of the above equation, which is very small compared to first term, the equation takes the form:

$$\beta_{a,\infty} \approx \beta_{T,\infty} \tag{9}$$

which is a valid assumption used in the present calculation.

Poisson's ratio can be obtained from the knowledge of the bulk modulus $k_{T,\infty}$ and the modulus of rigidity $G_{T,\infty}$, which arise from the change in lattice spacing corresponding to the solid-like character of the liquid. The Poisson's ratio is given by [25,26],

$$\sigma = \frac{3A - 2}{6A + 2} \tag{10}$$

and

$$A = \frac{k_{T,\infty}}{G_{T,\infty}} = \frac{4}{3}\frac{1}{\gamma}$$

where γ is the principal specific heat ratio and is laisely responsible for the maximum accuracy in the values of the parameter A and hence the effective Debye temperature.

The pseudo-Grüneisen parameter has been defined in terms of sound velocity as

$$\Gamma = \frac{U^2\alpha}{C_p} \quad \text{and} \quad U^2 = \frac{1}{\beta_s\rho} \tag{11}$$

and

$$\gamma = \frac{\beta_T}{\beta_s}$$

where α, C_p, β_T, β_s, γ and ρ are the thermal expansion coefficient, specific heat ratio at constant pressure, isothermal compressibility, adiabatic compressibility, specific heat ratio and density respectively. One can write

$$\Gamma = \frac{\beta_T}{\beta_s}\,\frac{T\alpha - 1}{T\alpha} \tag{12}$$

or

$$\Gamma = \frac{1}{T}\left[\frac{1}{\beta_s \gamma_p} - \frac{1}{\alpha}\right] \tag{13}$$

where γ_p is thermal pressure coefficient which is equal to

$$\gamma_p = \left(\frac{\partial P}{\partial T}\right)_v = \frac{\alpha}{\beta_T} \tag{14}$$

γ_p was evaluated by using Flory theory as;

$$P^* = \gamma_p T \tilde{V}^2 \tag{15}$$

where P^* is the characteristic pressure and $\tilde{V}$ is the reduced volume.

The values of the reduced and characteristic parameters for both pure and binary systems were calculated according to Flory theory.

For pure components

$$\tilde{V} = \left(\frac{\alpha T}{3\alpha T + 3} + 1\right)^3 \tag{16}$$

$$P^* = \frac{\alpha}{\beta_T} T \tilde{V}^2 \tag{17}$$

and for liquid mixtures

$$\tilde{V} = \frac{V_m}{\sum_{i=1}^{j} x_i V_i^*} \tag{18}$$

where x_i is the mole fraction, V_i^* is the characteristic volume of the component i, V_m is the molar volume of the liquid mixture and the characteristic pressure of the liquid mixture can be expressed as

$$P^* = \left[\sum_{i=1}^{j} \Psi_i P_i^* - \left(\sum_{i}^{j} \Psi_i \theta_j X_{ij}\right)\right] \tag{19}$$

Here ψ_i and θ_i are the segment fraction and site fraction of the component i while X_{ij} is the interaction parameter. The segment fraction as per Flory theory has been

calculated from the relation;

$$\Psi_j = (1 - \Psi_j) = \frac{x_j}{x_j + (x_i v_i^*/v_j^*)} \tag{20}$$

and the site fraction is calculated from

$$\theta_j = (1 - \theta_j) = \frac{\Psi_j}{\Psi_j + \Psi_i (v_j^*/v_i^*)^{1/3}}. \tag{21}$$

The interaction parameter X_{ij} has been evaluated following Berthelot's approximation for mono-polar species and is expressed as

$$X_{ij} = P_i^* \left[1 - \left(v_j^*/v_i^* \right)^{1/6} \left(P_j^*/P_i^* \right)^{1/2} \right]^2. \tag{22}$$

It has been pointed out by several workers that the excess thermodynamic functions are sensitively dependent not only on the difference in intermolecular forces, but also on the difference in size of the molecule [27]. Excess pseudo-Grüneisen parameter can be defined by the equation,

$$\Gamma^{\mathrm{E}} = \Gamma_{\mathrm{m}} - \Gamma_{\mathrm{idl}} \tag{23}$$

where Γ_{m} and Γ_{idl} are pseudo-Grüneisen parameters of Pb–Sn molten liquid mixture and ideal pseudo-Grüneisen parameter of liquid mixture respectively. The latter Γ_{idl} say, can be defined as;

$$\Gamma_{\mathrm{idl}} = x_1 \Gamma_1 + x_2 \Gamma_2 \tag{24}$$

where x_1 and x_2 are the mole fractions and Γ_1 and Γ_2 are the pseudo-Grüneisen parameters of Pb and Sn respectively.

3. Results and discussion

Values of molar volume (V), collision factor (S), space filling factor (r_f), molecular radius, actual volume (B), observed sound velocity (U_{exp}), theoretical sound velocity deduced from collision factor theory (U_{theo}) and intermolecular free length (L_{f}) of pure Pb and Sn are listed in table 1. The critical temperature (T_{c}), molar volume (V_{m}), molar volume at absolute zero (V_0), molar available volume (V_{a}), surface area (y), intermolecular free length ($L_{f_{\mathrm{m}}}$), collision factor (S_{m}), space filling factor ($r_{f_{\mathrm{m}}}$), molecular radius, actual volume (B_{m}), observed sound velocity (U_{exp}), theoretical sound velocity (U_{theo}) and percentage deviation (Δ) of Pb–Sn molten binary mixture are listed in tables 2 and 3, respectively.

Values of the thermal expansion coefficient (α) heat capacity (C_{p}), specific heat ratio (γ), pseudo-Grüneisen parameter (Γ), density (ρ), Poisson's ratio (σ) and effective Debye temperature (θ) of pure Pb and Sn are presented in table 4, whereas density (ρ), Poisson's ratio (σ), effective Debye temperature (θ), thermal expansion coefficient (α), heat capacity (C_{p}), specific heat ratio (γ), ideal pseudo-Grüneisen parameter (Γ_{idl}), pseudo-Grüneisen parameter (Γ_{m}) and excess pseudo-Grüneisen parameter (Γ^{E}) of the Pb–Sn molten mixture are presented in table 5. In order to carry out our calculations,

Table 1. Parameters of the pure component at various temperatures.

Components	Temp., t(°C)	Molar volume, V(cm^3)	Collision factor (S)	Space filling factor (r_f)	Molecular radius (r_m/Å)	Actual volume / mole (B cm^3 mole^{-1})	Observed ultrasonic velocity, U_{exp}(m s^{-1})	Theoretical ultrasonic velocity, U_{theo}(m s^{-1})	Inter molecular free length (L_f)
Sn	400	17.3097	6.6586	0.2384	1.1752	4.1293	2425	2425.4	0.0665
	500	17.5316	6.3157	0.2375	1.1785	4.1566	2400	2399.9	0.0779
	600	17.7079	6.2695	0.2368	1.1811	4.1914	2375	2375.4	0.0879
	700	17.8910	6.2410	0.2360	1.1872	4.2217	2365	2356.6	0.1020
Pb	400	19.7351	4.7332	0.2377	1.2296	4.6909	1800	1800.1	0.0964
	600	20.1441	4.6020	0.2356	1.2345	4.7467	1735	1734.8	0.1316
	700	204172	4.5406	0.2347	1.2384	4.7916	1705	1705.5	0.1508

Table 2. Parameters of the Pb–Sn molten binary mixtures.

Components (%)	Critical temp., T_c(°C)	Absolute temp., T(K)	Molar volume, V_m(c.c.)	Molar volume at absolute zero, V_0(c.c.)	Molar available volume, V_a(c.c.)	Surface area /mole ($Y \times 10^{-9}$ cm^2)	Inter molecular free length (L_f)
10	4115.00	673	17.5522	17.0341	0.9375	2.7040	0.0693
		873	17.9515	16.9994	1.2606	2.7004	0.0933
		973	18.1436	17.0524	1.4375	2.7059	0.1006
20	4008.33	673	17.7948	17.5697	0.9960	2.7604	0.0721
		873	18.1951	17.5621	1.3431	2.7596	0.0973
		973	18.3962	17.5634	1.5279	2.7598	0.1107
30	3901.67	673	18.0373	180268	1.0536	2.8084	0.0750
		873	18.4387	17.9478	1.4169	2.8002	0.1012
		973	18.6489	18.1351	1.6298	2.8196	0.1156
38	3815.27	673	18.2338	18.3616	1.1007	2.8430	0.0774
		873	18.6361	18.3662	1.4889	2.8435	0.1047
		973	18.8535	18.3861	1.6978	2.8456	0.1193
45	3741.66	673	18.4011	18.5116	1.1346	2.8582	0.0794
			18.8042	18.5508	1.5391	2.8622	0.1076
		873					
			19.0278	18.5516	1.7549	2.8632	0.1226
		973					
60	3581.67	673	18.7649	18.6620	1.2024	2.8737	0.0837
		873	19.1696	18.7486	1.6391	2.8826	0.1137
		973	19.4067	18.7123	1.8668	2.8788	0.1297
80	3368.33	673	19.2500	18.5597	1.2835	2.8632	0.0897
		873	19.6568	18.5319	1.7452	2.8603	0.1220
		973	19.9119	18.5637	1.9989	2.8636	0.1396

Table 3. Parameters of the Pb–Sn molten binary mixtures.

Components (%)	Temp., T(°C)	Collision factor (S_m)	Space filling factor (r_{fm})	Molecular radius (γ_m/Å)	Actual volume, B_m (cm^3 mole)	Expl ultrasonic velocity, U_{exp} ($m\,s^{-1}$)	Theo ultrasonic velocity, U_{theo} ($m\,s^{-1}$)	% age deviation (Δ)
10	673	6.1961	0.2383	1.1930	4.2834	2360.0	2362.2	0.01
	873	6.1028	0.2366	1.1964	4.3211	2320.0	2310.6	0.40
	973	6.0709	02358	1.2004	4.3636	2305.0	2302.7	0.10
20	673	6.0335	0.2382	1.2061	4.4266	2315.0	2299.6	0.66
	873	5.9360	0.2365	1.2105	4.452	2270.0	2246.3	1.0
	973	5.9009	0.2357	1.2131	4.5041	2250.0	2225.2	1.1
30	673	5.8710	0.2381	1.2170	4.5493	2235.0	2236.9	0.09
	873	5.7693	0.2364	1.2200	4.5813	2190.0	2182.1	0.36
	973	5.7309	0.2355	1.2269	4.6599	2170.0	2159.8	0.47
38	673	5.7393	0.2381	1.225	4.6414	2200.0	2186.3	0.62
	873	5.6342	0.2363	1.2303	4.6988	2150.0	2130.3	0.93
	973	5.5933	0.2354	1.2335	4.7375	2125.0	2107.0	0.85
45	673	5.6272	0.2380	1.2268	4.6583	2175.0	2143.1	1.47
	873	5.5191	0.2362	1.2351	4.7537	2120.0	2085.4	1.63
	973	5.4760	0.2353	1.2382	4.7899	2095.0	2062.0	1.57
60	673	5.3836	0.2379	1.2331	4.7311	2085.0	2049.5	1.73
	873	5.2690	0.2361	1.2406	4.8168	2030.0	1990.0	1.97
	973	5.2208	0.2352	1.2428	4.8436	2005.0	1964.3	2.03
80	673	5.0583	0.2378	1.2343	4.7443	1955.0	1924.6	1.55
	873	4.9355	0.2358	1.2380	4.7871	1895.0	1892.4	1.72
	973	4.8807	0.2349	1.2422	4.8362	1865.0	1834.5	1.64

Table 4. Parameters of the pure components at various temperatures.

Comp.	Temp., $T(^\circ C)$	Thermal expansion coefficient ($\alpha \times 10^5\,K^{-1}$)	Heat capacity ($C_p \times 10^3\,cal\,mol^{-1}$)	Specific heat ratio (γ)	Ultrasonic velocity, $U_{exp}\,(m\,s^{-1})$	Pseudo-Grüneisen parameter (Γ)	Density, $\rho\,(gm\,cm^{-3})$	Poisson's ratio (σ)	Effective Debye temp., $\theta(^\circ K)$
Sn	400	8.8600	60.2	1.125	2425	2.0901	6.8513	0.1707	224.29
	500	8.9761	61.2	1.140	2400	2.0177	6.7817	0.1673	221.15
	600	8.9986	62.3	1.151	2375	1.9222	6.7027	0.1648	218.70
	700	9.0997	63.3	1.171	2365	1.9318	6.6340	0.1603	216.18
Pb	400	12.4533	33.5	1.240	1800	2.8636	10.4980	0.1450	159.19
	600	12.7670	34.3	1.300	1735	2.6916	10.3745	0.1391	155.12
	700	12.8993	34.7	1.325	1705	2.5894	10.2849	0.1321	152.20

Table 5. Parameters of the Pb-Sn molten binary mixtures.

Comp. (%)	Temp., T(°C)	Density, ρ(gm cm^{-3})	Poisson's ratio (σ)	Effective Debye temp., θ(°K)	Thermal expansion coefficient, $\alpha \times 10^5$(K^{-1})	Heat capacity ($C_p \times 10^3$ cal mol^{-1})	Specific heat ratio (γ)	Observed ultrasonic velocity, U(m s^{-1})	Ideal pseudo-Grüneisen parameter (Γ_{idl})	Pseudo-Grüneisen parameter (Γ_{mix})	Excess pseudo-Grüneisen parameter (Γ^E)
10	673	7.0146	0.1682	214.75	9.8008	57.5	1.1500	2360	2.1674	2.2741	0.11
	773	7.0242	0.1605	212.89	9.8793	59.5	1.1700	2340	2.1576	2.2260	0.06
	873	6.9846	01556	210.57	10.1721	60.4	1.1920	2320	1.9995	2.1621	0.16
	973	6.8944	0.1512	208.21	10.1861	54.8	1.2120	2305	1.9975	2.1390	0.40
20	673	7.3462	0.1621	209.12	10.2477	55.7	1.1630	2315	2.2448	2.3634	0.12
	773	7.2743	0.1574	206.54	10.3614	56.7	1.1840	2295	2.1079	2.2973	0.18
	873	7.2143	0.1530	203.64	10.4071	57.6	1.2040	2270	2.0760	2.2451	0.17
	973	7.1440	0.1481	201.09	10.4859	52.2	1.2260	2250	2.0633	2.2151	0.15
30	673	7.6118	0.1583	200.02	10.8838	53.4	1.1800	2235	2.3221	2.4574	0.13
	773	7.5490	0.1534	198.18	10.8495	53.9	1.2020	2215	2.2347	2.4086	0.17
	873	7.5000	0.1483	197.27	11.0427	54.7	1.2250	2190	2.1530	2.3339	0.18
	973	7.3482	0.1429	191.64	11.1502	50.0	1.2500	2170	2.0295	2.3043	0.18
38	673	7.8321	0.1573	198.02	10.8234	50.8	1.1830	2200	2.3870	2.5123	0.13
	773	7.7246	0.1554	192.18	10.9924	51.6	1.2060	2175	2.2467	2.4243	0.17
	873	7.7021	0.1492	191.32	11.0136	52.4	1.2300	2150	.2.2153	2.3921	0.18
	973	7.8001	0.1463	182.64	11.0869	48.2	1.2510	2125	2.1821	2.3267	0.14
45	673	8.0683	0.1550	192.73	11.0988	48.9	1.1950	2175	2.4387	2.6106	0.17
	773	8.0123	0.1496	187.20	11.1804	49.7	1.2190	2150	2.3740	2.5340	0.16
	873	7.8900	0.1442	186.27	11.2993	50.5	1.2440	2120	2.2684	2.4735	0.20
	973	7.8028	0.1390	180.64	11.4169	44.1	1.2680	2095	2.2272	2.4125	0.18
60	673	8.6478	0.1418	183.87	12.7843	44.8	1.2550	2085	2.5542	2.9638	0.41
	773	8.5380	0.1342	180.76	13.1812	45.5	1.2900	2060	2.7013	2.8462	0.14
	873	8.4259	01278	177.24	13.3438	46.1	1.3200	2030	2.3838	2.7569	0.36
	973	8.3475	0.1210	174.40	13.6071	38.9	1.3520	2005	2.2694	2.6587	0.33
80	673	9.5490	0.1429	172.52	12.5712	39.4	1.2500	1955	2.7084	2.9549	0.24
	773	9.4357	0.1364	169.08	12.7607	39.9	1.2800	1925	2.6147	2.8387	0.22
	873	9.3447	0.1297	165.81	12.9543	40.4	1.3110	1895	2.5377	2.7479	0.21
	973	9.2148	0.1236	162.33	13.0263	38.3	1.4400	1865	2.4578	2.9825	0.22

all the necessary data were taken from the literature [13,28]. A careful perusal of tables 2 and 3 reveals that the theoretical sound velocity for Pb–Sn molten binary system compares well with the experimental results. The minimum and maximum percentage error ranges from 0.01 to 2.03 and average percentage error is 1.02, which is very much comparable to the experimental findings and clearly indicates the success of the theory. Discrepancies have been attributed at some places at (700°C, 60% Pb), (600°C, Pb 80%) and (400°C, Pb 60%) which is due to very few active centres and of similar nature and structure of the molecules. These centres have strong range molecular forces and thus interactions play a role. Temperature variation, the reliability of experimental data and the empirical relation used may of course contribute to such discrepancies.

Effect of temperature and composition on intermolecular free length have been studied. The available volume that measures the free energy of the system also provides active sites increases gradually with the rise of temperature. A careful perusal of table 2 shows that the values of surface area per mole increases generally with the rise in temperature. Small increment in the values of surface area confirms a weak molecular interaction in the present system.

As evident from tables 4 and 5, the values of effective Debye temperature for all the pure and molten binary systems decreases as the temperature increases. The Debye temperature of the associated liquid [29] shows similar behaviour as observed in the present calculation. These similarities regarding the magnitude of molten salts are sufficient basis for the prediction of its values. At elevated temperatures, the values of effective Debye temperature are more affected due to atomic motions in molten mixtures. A perusal of table 5 reveals that the concentration dependence of the effective Debye temperature in binary molten mixtures is affected by the size of the second component. In all the systems under investigation, the values of θ increase on decreasing the mole concentration of the component having higher molecular weight. This behaviour has been explained on the basis of the hole theory. This affects the expansivity and compressibility of the systems.

Taking the vibrational frequency of a one-dimensional crystal into consideration, Knopoff *et al.* [10] showed that $d\Gamma/dT > 0$. The pseudo-Grüneisen parameter for Pb–Sn molten mixture decreases in regular order, but the rate $(d\Gamma/dT)/T$ is somewhat higher. The change of pseudo-Grüneisen parameter corresponds to the relative change in the molecular order. A close examination of table 5 reveals that the value of pseudo-Grüneisen parameter increases at constant temperature as the composition of Pb is increased in a regular manner. The general trend of the pseudo-Grüneisen parameter with temperature variation [30] is decreasing. A significant observation can be made regarding the intermolecular interactions by observing the values of the excess pseudo-Grüneisen parameter. Higher values, say 0.40 and 0.36, can be seen at 700°C Pb 10% and at 600°C Pb 60% respectively, which suggests strong interactions at these temperatures.

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