International Journal of Thermodynamics (IJoT)
ISSN 1301-9724 / e-ISSN 2146-1511
www.ijoticat.com

Vol. 18 (No. 3), pp. 150-159, 2015
doi: 10.5541/ijot.5000057147
Published online: September 1, 2015

Pseudo-Gruneisen Parameter and Internal Pressure of Binary Mixtures from Different Approaches

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Received 03 August 2014, Revised 13 February 2015, Accepted 27 February 2015

Abstract

Density and speed of sound were measured earlier by us for binary liquid mixtures formed by Benzonitrile, Chlorobenzene, Benzyl chloride and Benzyl alcohol with Benzene at temperature range 298.15 to 313.15 K and atmospheric pressure over the whole concentration range. Pseudo-Gruneisen parameter and internal pressure were derived from the measured values of density and ultrasonic velocity. These values were compared with the theoretical values obtained by the utilization of Flory theory, Ramaswamy and Anbananthan model and model suggested by Glinski to predict the behavior of weakly interacting liquids. The observed properties derived from measured data were fitted to Redlich-Kister polynomial relation to estimate the binary coefficients and standard errors. Excess Pseudo-Gruneisen parameter for these binary mixtures was computed to study the molecular interactions involved in the liquid systems.

Keywords**:** *Pseudo-Gruneisen parameter; internal pressure; Flory theory; Ramaswamy and Anbananthan model: Redlich-Kister.*

1. Introduction

Gruneisen parameter is an important tool to study the thermodynamic and other properties of solid crystalline lattice [1].The concept of anharmonicity of the lattice is characterized by the Gruneisen parameter. It is well established that liquids support a quasi-crystalline model for their structure, the lattice nature being increased at high pressure and low temperature and the Gruneisen parameter can be used to study them. Its pseudo counterpart has been found to be suitable to investigate the internal structures, clustering phenomenon and other quasi crystalline properties of liquids, liquid mixtures [2-5], liquefied gases [6], liquid metal alloys [7] and higher alkanes [2].

The role of internal pressure in liquid solution thermodynamics was recognized by Hildebrand [8]. The use of this property for a long time was qualitative but recently its usefulness has been explored for quantitative study of intermolecular forces. Pioneer attempts have been made by several workers [9-15] to study the significance and its correlation with other properties. The internal pressure can be computed by indirect methods as suggested by Suryanarayan. In recent past, substantial amount of work has been carried out by many workers to study the excess thermodynamic functions like excess internal pressure; excess energy of vaporization, excess pseudo-Gruneisen parameter is still in progress [16-20].

In the present investigation, Pseudo-Gruneisen parameter, excess pseudo-Gruneisen parameter and internal pressure were derived from the measured values of density and ultrasonic velocity [21] for binary liquid mixtures formed by Benzonitrile, Chlorobenzene, Benzyl chloride and Benzyl alcohol with Benzene at temperature range 298.15 to 313.15 K and atmospheric pressure over the whole concentration range. A comparison of experimental and theoretical speed of sound is provided in Table 1. These derived values of pseudo-Gruneisen parameters were compared with the theoretical values obtained by the utilization of Flory theory [22-23], Ramaswamy and Anbananthan model [24] and model suggested by Glinski [25] to predict the behavior of weakly interacting liquids. The observed properties derived from measured data were fitted to Redlich-Kister polynomial relation [26] to estimate the binary coefficients and standard errors.

2. Theoretical

2.1 Flory Theory

Assuming two body interactions, Flory evaluated the reduced and characteristic parameters of the liquid mixture from the reduced equation of state derived from the resulting partition function as;

$$\frac{\tilde{P}\tilde{V}}{\tilde{T}} = \frac{\tilde{V}^{1/3}}{\tilde{V}^{1/3}-1} - \frac{1}{\tilde{V}\tilde{T}} \qquad (1)$$

where, $\tilde{P} = \frac{P}{P^*}$, $\tilde{T} = \frac{T}{T^*}$, and $\tilde{V} = \frac{V}{V^*}$ that are reduced parameters and P^*,T^*,$V^*$are characteristic parameters. The coefficient of thermal expansion, α, is evaluated from the reduced equation of state as:

$$a_{Flory} = \frac{1}{T\left[\frac{1}{3\left(\tilde{V}^{\frac{1}{3}}-1\right)-1}\right]} \qquad (2)$$

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adiabatic and compressibility is given by,

$$\beta_s = \frac{1}{u^2 \rho} \tag{3}$$

where u and ρ are experimental speed of sound and density of binary liquid mixture. By combining above equations Pseudo-Greneisen parameter for liquid mixtures is obtained as;

$$\tau_{Flory} = \frac{1}{T}\left[\frac{1}{\beta_s} - \gamma_P - \frac{1}{a}\right] \tag{4}$$

where γ_P is the thermal pressure coefficient related to the following expression as;

$$\gamma_P = \frac{P^*}{T\tilde{V}^2} \tag{5}$$

γ_P is also related to internal pressure of the liquid mixture as;

$$P_{i_{Flory}} = T.\gamma_P \tag{6}$$

For observed value of Pesudo-Gruneisen parameter, γ_P is evaluated by the relation as;

$$\gamma_P = \frac{a}{\beta_T} \tag{7}$$

Thermal expansion coefficient, α and adiabatic compressibility, β_T are defined as;

$$a = (0.0191 \times \beta_T)^{1/4} \tag{8}$$

$$\beta_T = 1.71 \times 10^{-3}/T^{4/9} \times u^2 \times \rho^{4/3} \tag{9}$$

2.2 Ramswamy and Anbananthan Model

Ramswamy and Anbananthan [24] proposed the model based on the assumption of linearity of acoustic impedance with the mole fraction of components. Further it is assumed, that an equilibrium physical property such as viscosity, refractive index, surface tension etc which are based on linearity can also be predicted. Glinski [25] assumed that when solute is added to solvent the molecules interact according to

$$A+B \leftrightarrow AB \tag{10}$$

and the association constant, K_{as}, can be defined as;

$$K_{as} = \frac{[AB]}{[A][B]} \tag{11}$$

where [*A*] is amount of solvent and [*B*] is amount of solute in the liquid mixture.

By applying the condition of linearity with composition, peudo-Gruneisen parameter can be obtained as;

$$\Gamma_{cal} = x_A \Gamma_A + x_{AB} \Gamma_{AB} \tag{12}$$

where x_A, x_{AB}, Γ_A, Γ_{AB} and Γ_{cal} are the mole fraction of A, mole fraction of associate AB, peudo-Gruneisen parameter of A, peudo-Gruneisen parameter of associate AB and calculated peudo-Gruneisen parameter respectively. The associate AB cannot be obtained in its pure form. Following simplifications have been made in Eq. (12), firstly, molar concentration term should be replaced by activities for concentrated solution and second, the equilibrium reaction is not complete by definition; i.e. there are also molecules of non-associated component present in the liquid mixture even prevailing in the high solute content. The Eq. (12) takes the form,

$$\Gamma_{cal} = [x_A \Gamma_A + x_B \Gamma_B + x_{AB} \Gamma_{AB}] \tag{13}$$

The general idea of this model can be, however, exploited as;

$$K_{as} = \frac{[AB]}{(C_A - [AB])(C_B - [AB])} \tag{14}$$

where C_A and C_B are initial molar concentrations of the components. One can take any value of association constant, K_{as}, and calculate the equilibrium value of [AB] for every composition of the mixture. Replacing molar concentration by equimolar activities for concentrated solution, Eq. (14) becomes,

$$K_{as} = \frac{a_{AB}}{(a_A - a_{AB})(a_B - a_{AB})} \tag{15}$$

where a_A, a_B and a_{AB} are the activity of component A, Component B and associate, AB respectively.

Similarly, assuming any value of peudo-Gruneisen parameter for hypothetical pure component AB, the peudo-Gruneisen parameter of liquid mixture, Γ_{cal} can be calculated by substituting the value of association constant. Now, it is possible to compare the peudo-Gruneisen parameter calculated using Eq (14) with the experimental values. On changing both the adjustable parameters K_{as} and Γ_{AB} gradually, one can get different values of the sum of squares of deviations,

$$S = \Sigma (\Gamma_{obs} - \Gamma_{cal})^2 \tag{16}$$

where Γ_{obs} and Γ_{cal} are the observed and calculated peudo-Gruneisen parameter respectively.

The minimum value of *S* can be obtained theoretically by a pair of the fitted parameters. But we found that for some K_{as} and Γ_{AB}, the value of *S i*s high and changes rapidly, and for others, it is low and changes slowly when changing the fitted parameters. The condition which is prevailing in the process of adjustment is that the value of Γ_{AB} should not be much lower than the lowest Γ_{obs} of thesystem or much higher than the highest one. Quantitatively, it should be reasonable to accept the pair of adjustable parameters K_{as} and Γ_{AB} which has the physical sense and which reproduces the experimental physical property satisfactorily.

On inspecting the results obtained from Ramaswamy and Anbananthan model, Glisnki [25] suggested the equation assuming additivity with the volume fraction, ϕ of the components, the refined version of Natta and Baccaredda model [27] as,

$$\tau_{cal} = \frac{\tau_A \tau_B \tau_{AB}}{\phi_A \tau_B \tau_{AB} + \phi_B \tau_A \tau_{AB} + \phi_{AB} \tau_A \tau_B} \tag{17}$$

where Γ_{cal} is the theoretical peudo-Gruneisen parameter of binary liquid mixture, ϕ_A, ϕ_B are the volume fractions of component A and B and Γ_A, Γ_B and Γ_{AB} are the peudo-

Gruneisen parameter of components A, B and AB. The numerical procedure and determination of association constant, K_{as}, were similar to that described before and the advantage of this method as compared with the earlier one was that the data on densities of liquid mixture are not necessary except those of pure components needed to calculate the volume fractions.

Table 1. Comparison of Density with literature data for pure components at 293.15, 298.15, 303.15, 308.15 and 313.15 K.

Comp.	$\alpha.10^3$ K	$\beta_T.10^{12}$ Pa	V/cm^3 mole^{-1}	T/^{o}K	ρ_{exp}/ g.cm^{-3}	ρ_{lit}/ g.cm^{-3}
B	1.218023	94.6097	89.319	298.15	0.8732	0.8736 [32]
	1.21875	94.7791	89.936	303.15	0.8680	0.8683 [30]
	1.228696	97.1185	90.730	308.15	0.8653	-
	1.24239	100.402	91.132	313.15	0.8575	0.8576 [30]
BN	0.997994	52.0415	103.078	298.15	1.0003	1.0006 [30]
	1.008302	53.6709	103.565	303.15	0.9976	0.9978 [30]
	1.010971	54.0982	104.841	308.15	0.9941	-
	1.016819	55.0424	105.245	313.15	0.9919	-
CB	0.99078	50.9211	102.234	298.15	1.1004	1.1009 [30]
	0.997456	51.9574	102.747	303.15	1.0951	1.0955 [30]
	1.004295	53.0335	102.841	308.15	1.0922	1.0926 [33]
	1.026654	56.6551	104.013	313.15	1.0863	1.0878 [33]
BC	1.059322	62.2374	115.649	298.15	1.0899	-
	1.067131	63.6240	116.158	303.15	1.0890	1.0897 [31]
	1.070387	64.2080	116.662	308.15	1.0894	-
	1.074842	65.0132	116.892	313.15	1.0802	1.0806 [30]
BA	1.015504	54.8292	103.821	298.15	1.0412	1.0413 [30]
	1.021907	55.8729	104.241	303.15	1.0370	1.0376 [30]
	1.033784	57.8437	105.450	308.15	1.0372	-
	1.063372	62.9539	107.978	313.15	1.0366	-

B: Benzene, BN: Benzonitrile, CB: Chlorobenzene,
BC: Benzylchloride, BA: Benzyl alcohol

3. Result and Discussion

Relations between associations phenomenon in liquids were analyzed earlier [28] by considering van der Waals equation of state which was based only on simple averaged geometrical deviations without analyzing the system in terms of equilibrium. The association phenomenon has been related usually the deviation of different quantities from additivity. Ramaswami and Anbananthan derived the model based on the assumption of linearity of acoustic impedance with the mole fraction of components which was corrected [25] and tested [28] to predict the associational behavior. The quantities analyzed were refractive index, molar volume, viscosity, intermolecular free length. Prediction of ultrasonic velocity from this approach is our first attempt. The results of fittings obtained from the model were utilized properly. The basic doubt regarding this model except the assumption of linearity of ultrasonic velocity with mole fraction is that these liquids have poor affinity to form dimmers. The calculations were performed using a computer program which allows fittings easily both the adjustable parameters simultaneously or the parameters were changed manually.

We constructed the data sheet in a computer program with association constant K_{as} and $\Gamma_{A,B}$ as the fitted parameters. On changing these parameters, the equilibrium concentrations of species [A], [B] and [AB] will change and the pseudo-Gruneisen parameter can be computed. The difference between experimental and theoretical values for pseudo-Gruneisen parameter is used to obtain the sum of squares of deviation. It is assumed that in solution three associates instead of two are formed (A, B and AB). The values of pseudo-Gruneisen parameter in pure associate can be treated as a fitted one with the value of K_{as}.

The standard deviation $\Delta\Gamma$ can be represented mathematically by Redlich-Kister polynomial equation [26] for correlating the experimental data as;

$$y = x_i(1-x_1)\sum_{i=0}^{p} A_i(2x_1-1)^i \qquad (18)$$

where y refers to deviation in pseudo-Gruneisen parameter ,y, ($\Delta\Gamma$), x_1 is the mole fraction and A_i is the coefficient. The values of coefficients were determined by a multiple regression analysis based on the least square method and are summarized along with the standard deviations between the experimental and fitted values of the respective function in Table 2. The values of standard deviations lie between 4.7×10^{-2} –2.88×10^{-3} respectively.

Table 2. Coefficients of the Redlich-Kister Equation and Standard Deviations (δ) for Binary Liquid Mixtures at Various Temperatures.

	T	A_0	A_1	A_2	A_3	(δ)Std dev
Benzene+benzonitrile						
$\Delta\Gamma$	298.15	-0.4853	-0.1947	-0.0670	-0.1777	0.0087
	303.15	-0.5431	-0.1510	-0.0301	0.0189	0.0101
	308.15	-0.4878	-0.1228	0.1048	-0.3550	0.0146
	313.15	-0.4878	-0.4257	0.2020	0.8098	0.0175
Benzene +chlorobenzene						
$\Delta\Gamma$	298.15	-0.1620	-0.6209	-0.0742	-0.1910	0.0093
	303.15	-0.1578	-0.6764	0.0947	-0.0851	0.0108
	308.15	-0.1477	-0.6336	0.0536	-0.2525	0.0084
	313.15	-0.0582	-0.5425	0.3648	0.4125	0.0093
Benzene +benzalchloride						
$\Delta\Gamma$	298.15	-0.0712	-0.4388	0.5654	0.5991	0.0106
	303.15	-0.0499	-0.4774	0.3859	0.9171	0.0093
	308.15	-0.0930	-0.3418	1.4715	-0.6704	0.0166
	313.15	-0.6033	0.0513	0.5973	0.2723	0.0181
Benzene +benzylalcohol						
$\Delta\Gamma$	298.15	-0.2795	-0.0590	0.2292	-0.0715	0.0047
	303.15	-0.1239	0.0654	0.0832	-0.2021	0.0102
	308.15	-0.2467	-0.2752	0.3804	0.8708	0.0171
	313.15	-0.4120	-0.2075	0.3521	0.0070	0.0288

The absolute average percent deviations (AAPD) in pseudo-Gruneisen parameter obtained from different models are provided in Table 3. A care full perusal of Tables 3 and 4 indicate that associated processes provide fairly good

results as compared to non-associated. Higher deviation values in PFP model can be explained as the model was developed for non-electrolyte γ-meric spherical chain molecules and the system under investigation have interacting and associating properties. Moreover, the expression used for the computation of thermal expansion coefficient, α and adiabatic compressibility,β_T [29] are also empirical in nature.

Table 3. Comparison of Absolute Average Percent Deviation (AAPD) values obtained from various liquid state models at Kas = 0.001.

Temp.	Γ_{PFP}	Γ_{RS}	Γ_{GLI}	$Pi_{PFP}.10^{-9}$ /Pa	$Pi_{RS}.10^{-9}$ /Pa	$Pi_{GLI}.10^{-9}$ /Pa
Benzene+benzonitrile						
298.15	5.6253	2.3612	3.1674	6.0895	6.8990	9.1750
303.15	5.9932	2.4811	3.2363	6.0679	7.2426	9.3679
308.15	6.0657	2.4992	3.2886	5.5162	7.2920	9.5136
313.15	6.5190	2.2901	3.1461	5.5815	6.6788	9.0849
Benzene+chlorobenzene						
298.15	4.5532	2.2116	1.8829	3.4635	6.4735	5.5282
303.15	4.6353	2.0635	1.7560	3.4258	6.0485	5.1622
308.15	5.1246	2.3403	2.0551	3.7008	6.8400	6.0227
313.15	4.2779	1.0460	0.8236	3.6936	3.0973	2.4477
Benzene+benzalchloride						
298.15	3.2169	0.4036	0.3612	5.0055	1.2118	1.0883
303.15	3.4629	0.4447	0.4470	5.3942	1.3355	1.3083
308.15	3.9764	0.6653	0.6758	7.0755	2.0026	1.7852
313.15	5.3303	0.6265	0.6331	25.8374	1.8724	1.5333
Benzene+benzylalcohol						
298.15	4.2271	1.4890	0.4345	2.2318	4.3944	2.6666
303.15	4.1700	1.1617	0.5912	2.1800	3.4396	1.7878
308.15	4.3388	1.2700	0.5113	3.2411	3.7610	2.2523
313.15	6.7962	2.8875	0.8976	3.7908	8.3644	6.9699

Mixture data are presented in Table 4. Density and speed of sound of mixture data were taken from reference [21]. With the increase of mole fraction, the values of observed pseudo-Gruneisen parameter, observed internal pressure, theoretical pseudo-Gruneisen parameters and theoretical internal pressure obtained from all the models decrease at all temperatures whereas no regular trends are observed for excess pseudo-Gruneisen parameters. For excess properties, sign and magnitude are of much important to describe the molecular interactions involved in the liquid system. Negative values of excess observed pseudo-Gruneisen parameters, Γ^E_{Obs} for all the systems show the strong molecular interactions between the liquid components. Dispersion type interactions and structural effects arising from interstitial accommodation because of differences in molecular volumes and free volumes between liquid components contribute negative terms to Γ^E. The repulsive forces between the loan pair of electrons leads to positive values of Γ^E suggest the presence of weak interactions between the component molecules and the favoring packing of unlike molecules.

4. Conclusions

Conclusively, models assuming associated processes give more reliable results as compared to non-associated processes and helpful in deducing the internal structure of associates through the fitted values of pseudo-Gruneisen parameter and internal pressure in a hypothetical pure associate and observed dependence of concentration on composition of a mixture.

Acknowledgment:

Authors are very thankful to U.G.C., New Delhi for financial and Department of Chemistry, V.S.S.D. College, for cooperation.

Nomenclature

α	Thermal Expansion Coefficient / K
β_T	Isothermal Compressibility / Pa
γ_p	Thermal Pressure Coefficient K.Pa^{-1}
x	Mole fraction / gm.
Γ	Pseudo Gruneisen parameter
T	Absolute Temperature / ^{o}K
V	Molar Volume / cm^{3}mole^{-1}
A_0	Ist order Coefficients of the R-K Equation
A_1	2nd order Coefficients of the R-K Equation
A_2	3rd order Coefficients of the R-K Equation
A_3	4th order Coefficients of the R-K Equation
ϕ	Volume Fraction / cm^{3}mole^{-1}
M	Molecular Weight/g.
ρ	Density/g.cm^{-3}
P^*	Characteristic Pressure/Pa
T^*	Characteristic Temperature/^{o}K
V^*	Characteristic, Core Volume / cm^{3}mole^{-1}
P	Reduced Pressure / Pa
V	Reduced Volume / cm^{3}mole^{-1}
$\tilde{T}$	Reduced Temperature / ^{o}K
β_s	Isoentropic Compressibility / Pa
K_{as}	Association constant
X_{AB}	Mole fraction of associate / g.
C_A & C_B	Initial molar concentrations of A&B
S	Sum of squares of deviations
a_A,	Activity of component A / g.mole.lit^{-1}
a_B	Activity of component B / g.mole.lit^{-1}
a_{AB}	Activity of associate AB / g.mole.lit^{-1}
δ	Standard deviation
Pi	Internal pressure / Pa
$\Delta\Gamma$	Deviation in pseudo-Gruneisen parameter
u	Speed of sound / ms^{-1}
Γ^E	Excess pseudo-Gruneisen parameter

Subscript

PFP	Prigogine-Flory-Patterson
RS	Ramaswami &Anbananthan
GLI	Glinski
obs	Observed
cal	Calculated
lit	Literature
exp	Experimental

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Table 4. Pseudo-Gruniesen Values, Excess Pseudo-Gruniesen Values and Internal Pressures from Various Models for Binary Liquid Mixtures at Various Temperatures.

Benzene+benzonitrile

x_1	Γ_{Obs}	Γ_{PFP}	Γ_{RS}	Γ_{GLI}	Γ^E_{Obs}	Γ^E_{PFP}	Γ^E_{RS}	Γ^E_{GLI}	$Pi_{Obs}10^{-9}$ /Pa	Pi_{PFP} 10^{-9}	Pi_{RS} 10^{-9}	Pi_{GLI} 10^{-9}
298.15												
0.1681	-2.881	-2.8425	-2.8543	-2.8336	-0.047	-0.008	-0.020	0.000	5.5970	5.1590	5.4410	5.3230
0.3126	-2.861	-2.7622	-2.8015	-2.7716	-0.087	0.010	-0.028	0.001	5.4890	4.9060	5.1540	4.9900
0.4381	-2.827	-2.6911	-2.7541	-2.7215	-0.107	0.029	-0.033	-0.001	5.3090	4.6860	4.9050	4.7330
0.5481	-2.799	-2.6277	-2.7097	-2.6784	-0.125	0.046	-0.035	-0.004	5.1620	4.4920	4.6810	4.5210
0.6453	-2.772	-2.5715	-2.6736	-2.6456	-0.139	0.061	-0.040	-0.012	5.0200	4.3200	4.5020	4.3620
0.7318	-2.710	-2.5201	-2.6331	-2.6099	-0.114	0.076	-0.036	-0.013	4.7050	4.1680	4.3110	4.1980
0.8093	-2.675	-2.4747	-2.6035	-2.5857	-0.111	0.089	-0.039	-0.021	4.5300	4.0310	4.1720	4.0870
0.8792	-2.602	-2.4317	-2.5616	-2.5498	-0.067	0.103	-0.026	-0.014	4.1800	3.9080	3.9880	3.9330
0.9423	-2.557	-2.3908	-2.5129	-2.5070	-0.048	0.117	-0.004	0.001	3.9870	3.8010	3.7840	3.7570
303.15												
0.1681	-2.822	-2.7632	-2.7790	-2.7599	-0.065	-0.006	-0.022	-0.003	5.6160	5.2160	5.3620	5.2520
0.3126	-2.805	-2.6890	-2.7352	-2.7076	-0.103	0.012	-0.033	-0.006	5.5210	4.9710	5.1180	4.9650
0.4381	-2.779	-2.6232	-2.6945	-2.6645	-0.125	0.030	-0.040	-0.010	5.3780	4.7570	4.8990	4.7380
0.5481	-2.745	-2.5645	-2.6564	-2.6276	-0.133	0.047	-0.044	-0.015	5.1880	4.5700	4.7020	4.5500
0.6453	-2.732	-2.5131	-2.6288	-2.6030	-0.157	0.062	-0.053	-0.027	5.1170	4.4040	4.5580	4.4250
0.7318	-2.656	-2.4643	-2.5841	-2.5628	-0.114	0.077	-0.041	-0.020	4.7220	4.2560	4.3450	4.2380
0.8093	-2.606	-2.4202	-2.5421	-2.5259	-0.093	0.092	-0.029	-0.013	4.4750	4.1240	4.1510	4.0720
0.8792	-2.555	-2.3815	-2.5135	-2.5027	-0.069	0.104	-0.027	0.016	4.2220	4.0040	4.0200	3.9680
0.9423	-2.500	-2.3436	-2.4659	-2.4605	-0.038	0.118	-0.003	0.001	3.9800	3.8970	3.8150	3.7910
308.15												
0.1681	-2.755	-2.7035	-2.7253	-2.7057	-0.036	0.015	-0.006	0.013	5.5720	5.3290	5.3900	5.2740
0.3126	-2.750	-2.6291	-2.6835	-2.6552	-0.089	0.032	-0.021	0.006	5.5450	5.0720	5.1490	4.9880
0.4381	-2.715	-2.5616	-2.6332	-2.6026	-0.103	0.050	-0.021	0.009	5.3500	4.8480	4.8790	4.7100
0.5481	-2.686	-2.5036	-2.6018	-2.5722	-0.118	0.064	-0.033	-0.004	5.1850	4.6510	4.7080	4.5490
0.6453	-2.680	-2.4517	-2.5730	-2.5467	-0.150	0.077	-0.043	-0.017	5.1500	4.4770	4.5560	4.4170
0.7318	-2.598	-2.4030	-2.5301	-2.5083	-0.103	0.092	-0.034	-0.013	4.7100	4.3230	4.3450	4.2340
0.8093	-2.548	-2.3579	-2.4827	-2.4662	-0.083	0.106	-0.018	-0.001	4.4600	4.1840	4.1240	4.0420

0.8792	-2.505	-2.3198	-2.4593	-2.4482	-0.068	0.116	-0.022	-0.011	4.2420	4.0600	4.0120	3.9580
0.9423	-2.455	-2.3364	-2.4138	-2.4083	-0.043	0.075	-0.002	0.003	4.0120	3.5320	3.8120	3.7860
313.15												
0.1681	-2.709	-2.6376	-2.6709	-2.6495	-0.045	0.025	-0.007	0.013	5.6350	5.4100	5.4000	5.2710
0.3126	-2.690	-2.5620	-2.6201	-2.5894	-0.084	0.043	-0.014	0.016	5.5310	5.1430	5.1070	4.9300
0.4381	-2.669	-2.4962	-2.5827	-2.5495	-0.113	0.059	-0.026	0.006	5.4050	4.9100	4.8950	4.7080
0.5481	-2.618	-2.4366	-2.5394	-2.5075	-0.106	0.075	-0.027	0.004	5.1140	4.7050	4.6630	4.4890
0.6453	-2.620	-2.3852	-2.5139	-2.4855	-0.146	0.088	-0.040	-0.012	5.1200	4.5250	4.5230	4.3710
0.7318	-2.529	-2.3358	-2.4665	-2.4431	-0.090	0.103	-0.027	-0.004	4.6240	4.3640	4.2890	4.1680
0.8093	-2.482	-2.2924	-2.4312	-2.4133	-0.074	0.115	-0.022	-0.005	4.3820	4.2210	4.1170	4.0270
0.8792	-2.437	-2.2531	-2.3979	-2.3860	-0.057	0.127	-0.017	-0.005	4.1590	4.0920	3.9600	3.9010
0.9423	-2.357	-2.2163	-2.3589	-2.3530	-0.001	0.139	-0.003	0.002	3.7750	3.9760	3.7840	3.7550

Benzene+chlorobenzene

x_1	Γ_{Obs}	Γ_{PFP}	Γ_{RS}	Γ_{GLI}	Γ^E_{Obs}	Γ^E_{PFP}	Γ^E_{RS}	Γ^E_{GLI}	$Pi_{Obs}10^{-9}$ /Pa	Pi_{PFP} 10^{-9}	Pi_{RS} 10^{-9}	Pi_{GLI} 10^{-9}
298.15												
0.1808	-2.812	-2.8536	-2.7846	-2.7914	0.039	-0.002	0.067	0.060	5.2070	5.4520	5.0540	5.0920
0.3318	-2.783	-2.7664	-2.7433	-2.7539	0.001	0.017	0.040	0.030	5.0540	5.1570	4.8400	4.8970
0.4598	-2.749	-2.6896	-2.6951	-2.7074	-0.023	0.037	0.031	0.019	4.8910	4.9060	4.6040	4.6680
0.5697	-2.733	-2.6251	-2.6710	-2.6835	-0.055	0.052	0.006	-0.006	4.8030	4.6890	4.4830	4.5460
0.6651	-2.716	-2.5675	-2.6405	-2.6522	-0.082	0.067	-0.006	-0.017	4.7230	4.5010	4.3380	4.3960
0.7487	-2.691	-2.5158	-2.6073	-2.6175	-0.094	0.081	-0.010	-0.020	4.6040	4.3350	4.1870	4.2360
0.8225	-2.674	-2.4709	-2.5845	-2.5926	-0.110	0.093	-0.020	-0.028	4.5250	4.1890	4.0830	4.1210
0.8882	-2.602	-2.4289	-2.5502	-2.5558	-0.067	0.105	-0.015	-0.021	4.1800	4.0590	3.9350	3.9610
0.9470	-2.554	-2.3894	-2.5052	-2.5080	-0.046	0.118	0.002	0.000	3.9780	3.9430	3.7510	3.7630
303.15												
0.1808	-2.742	-2.7838	-2.7219	-2.7281	0.052	0.010	0.072	0.066	5.1520	5.2880	5.0370	5.0720
0.3318	-2.722	-2.7009	-2.6839	-2.6936	0.006	0.028	0.045	0.035	5.0470	5.0130	4.8360	4.8890
0.4598	-2.690	-2.6282	-2.6414	-2.6526	-0.017	0.045	0.032	0.021	4.8880	4.7800	4.6230	4.6820
0.5697	-2.681	-2.5671	-2.6202	-2.6317	-0.055	0.059	0.006	-0.005	4.8390	4.5790	4.5120	4.5720
0.6651	-2.670	-2.5133	-2.5993	-2.6101	-0.085	0.071	-0.014	-0.025	4.7770	4.4050	4.4070	4.4620
0.7487	-2.638	-2.4633	-2.5612	-2.5705	-0.089	0.085	-0.012	-0.021	4.6260	4.2530	4.2310	4.2770

0.8225	-2.605	-2.4187	-2.5253	-2.5326	-0.088	0.098	-0.008	-0.015	4.4700	4.1190	4.0700	4.1050
0.8882	-2.555	-2.3803	-2.5033	-2.5084	-0.066	0.108	-0.014	-0.019	4.2220	4.0010	3.9710	3.9950
0.9470	-2.499	-2.3426	-2.4600	-2.4625	-0.036	0.120	0.003	0.001	3.9760	3.8950	3.7890	3.8010
308.15												
0.1808	-2.685	-2.7134	-2.6609	-2.6666	0.052	0.024	0.076	0.071	5.1530	5.3630	5.0130	5.0460
0.3318	-2.664	-2.6317	-2.6237	-2.6326	0.008	0.041	0.049	0.040	5.0430	5.0820	4.8130	4.8630
0.4598	-2.628	-2.5589	-2.5735	-2.5837	-0.010	0.059	0.045	0.035	4.8620	4.8430	4.5610	4.6160
0.5697	-2.628	-2.5001	-2.5611	-2.5716	-0.056	0.071	0.010	0.000	4.8530	4.6390	4.4910	4.5460
0.6651	-2.618	-2.4471	-2.5394	-2.5492	-0.087	0.084	-0.008	-0.01	4.8030	4.4620	4.3800	4.4310
0.7487	-2.593	-2.3983	-2.5040	-2.5125	-0.097	0.097	-0.008	-0.01	4.6800	4.3070	4.2130	4.2560
0.8225	-2.547	-2.3539	-2.4637	-2.4704	-0.082	0.110	0.000	-0.006	4.4550	4.1710	4.0310	4.0640
0.8882	-2.505	-2.3170	-2.4477	-2.4523	-0.069	0.119	-0.011	-0.016	4.2420	4.0500	3.9560	3.9780
0.9470	-2.455	-2.2805	-2.4083	-2.4106	-0.043	0.130	0.003	0.001	4.0120	3.9430	3.7860	3.7970
313.15												
0.1808	-2.602	-2.6078	-2.5762	-2.5814	0.051	0.046	0.078	0.072	4.9990	5.2980	4.8460	4.8760
0.3318	-2.574	-2.5327	-2.5407	-2.5489	0.020	0.062	0.054	0.046	4.8440	5.0350	4.6570	4.7020
0.4598	-2.541	-2.4677	-2.5061	-2.5155	0.003	0.077	0.038	0.029	4.6710	4.8110	4.4790	4.5300
0.5697	-2.519	-2.4123	-2.4841	-2.4937	-0.017	0.089	0.017	0.007	4.5540	4.6180	4.3650	4.4160
0.6651	-2.510	-2.3646	-2.4695	-2.4785	-0.046	0.099	-0.005	-0.014	4.5080	4.4510	4.2880	4.3350
0.7487	-2.465	-2.3192	-2.4324	-2.4402	-0.034	0.112	-0.001	-0.009	4.2830	4.3050	4.1140	4.1530
0.8225	-2.432	-2.2799	-2.4062	-2.4123	-0.030	0.122	-0.004	-0.010	4.1250	4.1770	3.9920	4.0230
0.8882	-2.378	-2.2449	-2.3825	-2.3867	-0.001	0.131	-0.006	-0.010	3.8620	4.0630	3.8840	3.9050
0.9470	-2.351	-2.2121	-2.3506	-2.3527	0.001	0.141	0.002	0.001	3.7480	3.9610	3.7450	3.7550

Benzene+benzalchloride

x_1	Γ_{Obs}	Γ_{PFP}	Γ_{RS}	Γ_{GLI}	Γ^E_{Obs}	Γ^E_{PFP}	Γ^E_{RS}	Γ^E_{GLI}	$Pi_{Obs}10^{-9}$/Pa	Pi_{PFP} 10^{-9}	Pi_{RS} 10^{-9}	Pi_{GLI} 10^{-9}
298.15												
0.1988	-2.682	-2.6829	-2.6777	-2.6818	0.046	0.046	0.051	0.047	4.5150	4.6410	4.4920	4.5120
0.3583	-2.677	-2.6243	-2.6634	-2.6696	0.002	0.055	0.016	0.010	4.4920	4.4640	4.4210	4.4520
0.4894	-2.646	-2.5726	-2.6341	-2.6412	-0.006	0.067	0.006	-0.001	4.3510	4.3150	4.2890	4.3240
0.5982	-2.633	-2.5300	-2.6206	-2.6276	-0.026	0.076	-0.013	-0.020	4.2870	4.1870	4.2270	4.2610
0.6907	-2.595	-2.4910	-2.5917	-2.5981	-0.016	0.087	-0.013	-0.019	4.1190	4.0770	4.1030	4.1330
0.7701	-2.561	-2.4568	-2.5643	-2.5698	-0.007	0.097	-0.009	-0.015	3.9750	3.9810	3.9880	4.0130

0.8392	-2.535	-2.4273	-2.5433	-2.5476	-0.002	0.106	-0.009	-0.014	3.8660	3.8970	3.9000	3.9200
0.8993	-2.494	-2.3994	-2.5112	-2.5141	0.020	0.115	0.003	0.002	3.6960	3.8230	3.7720	3.7850
0.9526	-2.476	-2.3765	-2.4953	-2.4968	0.022	0.122	0.003	0.002	3.6250	3.7560	3.7080	3.7150
303.15												
0.1988	2.6407	-2.6175	-2.6280	-2.6311	0.031	0.054	0.044	0.041	4.5890	4.7170	4.5230	4.5390
0.3583	-2.615	-2.5609	-2.6015	-2.6062	0.010	0.065	0.024	0.019	4.4720	4.5450	4.3990	4.4220
0.4894	-2.597	-2.5134	-2.5808	-2.5861	-0.009	0.074	0.007	0.002	4.3860	4.4000	4.3030	4.3300
0.5982	-2.580	-2.4740	-2.5723	-2.5776	-0.024	0.082	-0.015	-0.021	4.3030	4.2760	4.2620	4.2880
0.6907	-2.542	-2.4372	-2.5427	-2.5475	-0.012	0.092	-0.013	-0.017	4.1300	4.1690	4.1320	4.1560
0.7701	-2.514	-2.4051	-2.5158	-2.5198	-0.008	0.101	-0.009	-0.013	4.0120	4.0760	4.0170	4.0360
0.8392	-2.482	-2.3767	-2.4904	-2.4935	0.004	0.110	-0.003	-0.006	3.8710	3.9940	3.9100	3.9250
0.8993	-2.452	-2.3518	-2.4686	-2.4708	0.017	0.117	0.000	-0.001	3.7450	3.9220	3.8200	3.8300
0.9526	-2.426	-2.3299	-2.4502	-2.4513	0.027	0.124	0.003	0.003	3.6390	3.8570	3.7450	3.7500
308.15												
0.1988	2.4935	-2.5468	-2.4821	-2.4852	0.131	0.077	0.142	0.139	4.1800	4.8140	4.1220	4.1380
0.3583	-2.568	-2.5031	-2.5549	-2.5599	0.009	0.074	0.022	0.017	4.5060	4.6320	4.4360	4.4620
0.4894	-2.549	-2.4536	-2.5249	-2.5305	-0.010	0.085	0.014	0.008	4.4200	4.4790	4.2960	4.3240
0.5982	-2.510	-2.4112	-2.4974	-2.5029	-0.003	0.095	0.009	0.004	4.2360	4.3480	4.1700	4.1980
0.6907	-2.487	-2.3758	-2.4817	-2.4868	-0.008	0.103	-0.002	-0.007	4.1290	4.2350	4.0990	4.1240
0.7701	-2.468	-2.3449	-2.4666	-2.4709	-0.012	0.111	-0.010	-0.014	4.0410	4.1370	4.0310	4.0520
0.8392	-2.421	-2.3159	-2.4386	-2.4420	0.014	0.120	-0.002	-0.005	3.8290	4.0510	3.9110	3.9270
0.8993	-2.389	-2.2907	-2.4162	-2.4185	0.029	0.127	0.002	-0.000	3.6900	3.9750	3.8160	3.8270
0.9526	-2.389	-2.2604	-2.4227	-2.4239	0.013	0.142	-0.020	-0.021	3.6850	3.9940	3.8410	3.8460
313.15												
0.1988	-2.432	-2.4867	-2.4245	-2.4282	-0.070	-0.124	-0.062	-0.065	4.1350	4.8990	4.0920	4.1110
0.3583	-2.512	-2.9067	-2.4960	-2.5019	-0.155	-0.550	-0.139	-0.145	4.4900	6.5220	4.4040	4.4350
0.4894	-2.497	-2.7649	-2.4662	-2.4728	-0.145	-0.413	-0.114	-0.121	4.4250	5.9770	4.2610	4.2960
0.5982	-2.464	-2.6438	-2.4446	-2.4512	-0.117	-0.296	-0.097	-0.103	4.2630	5.5200	4.1600	4.1930
0.6907	-2.437	-2.5385	-2.4187	-2.4247	-0.093	-0.194	-0.074	-0.080	4.1380	5.1340	4.0420	4.0720
0.7701	-2.406	-2.4478	-2.4011	-2.4062	-0.065	-0.106	-0.060	-0.065	3.9890	4.8040	3.9620	3.9880
0.8392	-2.379	-2.3680	-2.3798	-2.3837	-0.041	-0.029	-0.041	-0.045	3.8670	4.5190	3.8680	3.8880
0.8993	-2.345	-2.2984	-2.3613	-2.3640	-0.009	0.037	-0.025	-0.027	3.7120	4.2730	3.7880	3.8010
0.9526	-2.324	-2.2374	-2.3452	-2.3465	0.009	0.096	-0.010	-0.012	3.6210	4.0580	3.7190	3.7260

Benzene+benzylalcohol

x_1	Γ_{Obs}	Γ_{PFP}	Γ_{rs}	Γ_{Gli}	Γ^E_{Obs}	Γ^E_{PFP}	Γ^E_{RS}	Γ^E_{GLI}	Pi_{Obs}	Pi_{PFP} 10^{-9}	Pi_{RS} 10^{-9}	Pi_{GLI} 10^{-9}
298.15												
0.1749	-2.822	-2.7947	-2.7911	-2.8022	-0.018	0.008	0.012	0.001	5.2570	5.0000	5.0860	5.1470
0.3229	-2.791	-2.7208	-2.7524	-2.7702	-0.045	0.025	-0.006	-0.024	5.0920	4.7690	4.8830	4.9780
0.4498	-2.768	-2.6556	-2.7140	-2.7353	-0.071	0.041	-0.017	-0.038	4.9750	4.5690	4.6900	4.8010
0.5598	-2.721	-2.5966	-2.6669	-2.6890	-0.066	0.057	-0.012	-0.034	4.7440	4.3950	4.4660	4.5780
0.6561	-2.686	-2.5450	-2.6300	-2.6511	-0.068	0.072	-0.012	-0.033	4.5750	4.2430	4.2940	4.3990
0.7410	-2.635	-2.4982	-2.5896	-2.6082	-0.050	0.086	-0.005	-0.023	4.3360	4.1080	4.1140	4.2040
0.8166	-2.585	-2.4555	-2.5482	-2.5632	-0.029	0.099	0.007	-0.008	4.1100	3.9880	3.9370	4.0060
0.8841	-2.554	-2.4194	-2.5269	-2.5375	-0.025	0.109	0.002	-0.008	3.9680	3.8810	3.8430	3.8920
0.9450	-2.516	-2.3857	-2.4995	-2.5050	-0.010	0.119	0.006	0.001	3.8040	3.7840	3.7290	3.7540
303.15												
0.1749	-2.763	-2.7289	-2.7401	-2.7504	-0.014	0.020	0.008	-0.001	5.2590	5.0890	5.1260	5.1840
0.3229	-2.735	-2.6579	-2.6986	-2.7152	-0.011	0.066	0.025	0.009	5.1100	4.8600	4.9070	4.9970
0.4498	-2.710	-2.5959	-2.6616	-2.6814	-0.039	0.075	0.009	-0.010	4.9790	4.6630	4.7160	4.8220
0.5598	-2.663	-2.5403	-2.6204	-2.6410	-0.038	0.084	0.004	-0.016	4.7380	4.4910	4.5150	4.6220
0.6561	-2.616	-2.4905	-2.5796	-2.5992	-0.031	0.093	0.004	-0.014	4.5090	4.3400	4.3220	4.4220
0.7410	-2.558	-2.4438	-2.5255	-2.5427	-0.009	0.105	0.023	0.006	4.2440	4.2060	4.0810	4.1650
0.8166	-2.525	-2.4051	-2.4998	-2.5137	-0.008	0.112	0.017	0.003	4.0910	4.0870	3.9650	4.0320
0.8841	-2.501	-2.3711	-2.4813	-2.4912	-0.012	0.117	0.007	-0.002	3.9750	3.9810	3.8820	3.9280
0.9450	-2.472	-2.3402	-2.4637	-2.4688	-0.008	0.123	0.000	-0.004	3.8440	3.8860	3.8030	3.8270
308.15												
0.1749	-2.698	-2.7045	-2.6736	-2.6837	-0.016	-0.021	0.009	-0.001	5.2220	5.3390	5.0770	5.1340
0.3229	-2.674	-2.5804	-2.6341	-2.6503	-0.044	0.049	-0.004	-0.020	5.0920	4.8960	4.8640	4.9540
0.4498	-2.651	-2.5211	-2.5979	-2.6172	-0.066	0.063	-0.013	-0.032	4.9710	4.7010	4.6760	4.7810
0.5598	-2.594	-2.4677	-2.5562	-2.5763	-0.049	0.077	-0.010	-0.030	4.6730	4.5310	4.4690	4.5760
0.6561	-2.553	-2.4206	-2.5206	-2.5397	-0.042	0.090	-0.009	-0.028	4.4680	4.3810	4.2970	4.3960
0.7410	-2.511	-2.3772	-2.4773	-2.4942	-0.030	0.103	0.003	-0.013	4.2700	4.2490	4.0990	4.1830
0.8166	-2.467	-2.3381	-2.4372	-2.4507	-0.013	0.115	0.016	0.003	4.0680	4.1310	3.9200	3.9860
0.8841	-2.436	-2.3049	-2.4142	-2.4238	-0.006	0.125	0.015	0.006	3.9250	4.0260	3.8180	3.8630
0.9450	-2.378	-2.2751	-2.3946	-2.3997	0.030	0.133	0.013	0.008	3.6540	3.9320	3.7310	3.7550
313.15												
0.1749	-2.607	-2.5308	-2.5887	-2.5974	-0.018	0.058	0.001	-0.008	5.0180	5.0090	4.9080	4.9580
0.3229	-2.623	-2.4727	-2.5585	-2.5725	-0.079	0.070	-0.015	-0.029	5.1110	4.8090	4.7430	4.8220
0.4498	-2.547	-2.4205	-2.5205	-2.5372	-0.044	0.083	-0.016	-0.033	4.6970	4.6360	4.5480	4.6390
0.5598	-2.605	-2.3744	-2.4865	-2.5040	-0.136	0.095	-0.017	-0.034	5.0370	4.4840	4.3780	4.4700
0.6561	-2.569	-2.3321	-2.4450	-2.4615	-0.129	0.107	-0.005	-0.021	4.8490	4.3510	4.1800	4.2650
0.7410	-2.487	-2.2926	-2.3948	-2.4094	-0.074	0.120	0.018	0.0037	4.4310	4.2330	3.9530	4.0250
0.8166	-2.435	-2.2589	-2.3625	-2.3742	-0.046	0.130	0.027	0.0154	4.1740	4.1270	3.8080	3.8650
0.8841	-2.401	-2.2283	-2.3304	-2.3387	-0.032	0.140	0.038	0.0299	4.0130	4.0330	3.6690	3.7080
0.9450	-2.369	-2.2015	-2.3068	-2.3111	-0.019	0.148	0.042	0.0385	3.8650	3.9470	3.5680	3.5880