Theoretical evaluation of internal pressure in ternary and sub-binary liquid mixtures at various temperatures

J. Madhumitha, N. Santhi*, G. Alamelumangai, M. Emayavaramban
Department of Chemistry, Government Arts College, C.Mutlur, Chidambaram – 608102, India

*E-mail address: nsaanthi@gmail.com

ABSTRACT

The Internal pressures of Ternary and their sub-Binary liquid mixtures of benzene(1) + hexane(2) + sec-butyl alcohol(3) were calculated using density, velocity and molar refraction from the temperature range of 303.15K-318.15K. For the Binary liquid mixtures, the Experimental Internal pressure values were correlated through an equation proposed by Andiappan et.al. For the Ternary liquid mixture, the Experimental Internal pressure values were correlated through an equation proposed by us. The Experimental values and the Theoretical values are in close agreement with each other.

Keywords:
Ternary liquid mixtures; sub-binary liquid mixtures; benzene; hexane; sec-butyl alcohol

1. INTRODUCTION

The Internal pressure is the cohesive force, which is a resultant of forces of attraction and forces of repulsion between molecules in a liquid, and considerable importance can be gained by simply observing and comparing internal pressure-volume curves for pure liquids. The term a/V² in Vander waals equation being the measure of attractive force of the molecule is called the cohesive or internal pressure. The intermolecular forces give a liquid its cohesion and it creates a pressure of thousand to ten thousand atmospheres within a liquid. Cohesion creates a pressure within a liquid of between 10³ to 10⁴ atmospheres. The internal pressure thus depends markedly on the molar volume, and also on the external pressure. It should be emphasized that the internal pressure is also sensitive to the repulsive component in a liquid.

Non-polar liquids have low internal pressures of the order of 2000 to 3000 atm; polar liquids have somewhat larger values, hydrogen bonding still increasing the value, water having a value around twenty thousand atmospheres. The importance of internal pressure in understanding the properties of liquids and the full potential of internal pressure as a structural probe did become apparent with the Pioneering work of Hildebrand.
excellent article Dack\textsuperscript{7} reviewed the importance of solvent internal pressure and cohesion to solution phenomena. The internal pressure is a single factor which varies due to all type of solvent-solute, solute-solute and solvent-solvent interactions. The Various thermodynamic properties and molecular interactions involving self-associated alcohols and phenols helps in understanding the inter and intramolecular interactions.

Stavely et al\textsuperscript{8} compared the internal pressure of individual liquid components and predicted the interaction in liquid mixtures. Richards\textsuperscript{9} pointed out the importance of internal pressure in understanding the physical properties of liquids and solids. An extensive list of values of internal pressure for liquids has been given by Allen et al\textsuperscript{10}.

2. MATERIALS AND METHODS

The chemicals used in the present work are Analar grade and purified by the standard methods\textsuperscript{11}. The purity of samples are checked out by measuring their density, boiling point and refractive index (Table 1).

| Liquid          | Density(g/cm\textsuperscript{3}) | Boiling point | Refractive Index(n) |
|-----------------|----------------------------------|---------------|---------------------|
| n-hexane        | 0.6493                           | 0.6548        | 68.70               |
| Benzene         | 0.8675                           | 0.8736        | 80.10               |
| sec-butyl alcohol | 0.7988                          | 0.8026        | 99.50               |

Ultrasonic velocities of pure liquids and liquid mixtures from the temperature range of 308K to 318K were measured using Ultrasonic Interferometer operating at 3 MHz. The density was determined at the experimental temperatures using 10 ml capacity specific gravity bottle immersed in a thermostatic bath (accuracy + 0.01 °C). The volume of the bottle at the experimental temperature, viz. 308K-318K was ascertained using doubly distilled water.
3. THEORY AND CALCULATION

Srivastava and Berkowitz Equation\textsuperscript{12} equation was used to compute internal pressure from the measurement of ultrasonic velocity, density and refractive index.

Srivastava and Berkowitz Equation is

\[ U \sqrt{D} = K' \sqrt{(Rm) \pi_i} \] (1)

Where:

- \( U \) = Ultrasonic velocity
- \( D \) = Density
- \( Rm \) = Molar refraction
- \( K' \) = distinctly structure dependent Constant
- \( \pi_i \) = Internal pressure

If the data for ultrasonic velocity, density, Molar refraction, internal pressure of pure liquids are known, the constant (\( K' \)) can be determined using equation (1).

Srivastava and Berkowitz Equation for the Binary Liquid Mixtures is

\[
\left( \pi_i \right)_{12} = \frac{U_{12} \sqrt{D_{12}}}{(x_1 K'_1 + x_2 K'_2) \sqrt{(Rm)_{12}}} \] (2)

Where

- \( U_{12} \) = Ultrasonic velocity of the binary mixture
- \( D_{12} \) = Density of the binary mixture
- \( (Rm)_{12} \) = Molar refraction of the binary mixture
- \( x_1 \) and \( x_2 \) are Mole fractions of components (1) and (2)
- \( K'_1 \) and \( K'_2 \) are distinctly structure dependent constants
- \( (\pi_i)_{12} \) = Internal pressure of the binary mixture
Evaluation of $R_m$:

$$R_m = \left( \frac{n^2 - 1}{n^2 + 2} \right) \left( \frac{x_1 m_1 + x_2 m_2}{d} \right)$$ \hspace{1cm} (3)

Where

- $n =$ refractive index
- $x_1$ and $x_2$ are Mole fractions of components (1) and (2)
- $m_1$ and $m_2$ are Molecular weight of components (1) and (2)
- $d =$ density

The Andiappan et al\textsuperscript{13} equation was used to calculate the theoretical internal pressure of binary liquid mixtures.

Andiappan et al equation is

$$\log \pi_i = x_1 \log \pi_1 + x_2 \log \pi_2 - \beta x_1 x_2$$ \hspace{1cm} (4)

Where $x_1$ and $\pi_1$ are the mole fraction and internal pressure of the component 1 and $x_2$ and $\pi_2$ are the mole fraction and internal pressure of the component 2. The equation (4) containing only one constant $\beta$, has been employed for correlating the experimental data.

The experimentally determined internal pressure data for all the three binary systems have been correlated through equation (4) at temperatures 303.15K to 318.15K (Table 3). Constant $\beta$ has been determined through a least square method at all the temperatures.

Srivastava and Berkowitz equation for the Ternary liquid mixture is written as

$$\left( \pi_i \right)_{123} = \frac{U_{123} \sqrt{D_{123}}}{(x_1 K'_1 + x_2 K'_2 + x_3 K'_3) \sqrt{(Rm)_{123}}}$$ \hspace{1cm} (5)

Where

- $U_{123} =$ Ultrasonic velocity of the ternary mixture
- $D_{123} =$ Density of the ternary mixture
- $(Rm)_{123} =$ Molar refraction of the ternary mixture
- $x_1$, $x_2$, and $x_3$ are Mole fractions of components (1), (2) and (3)
- $K'_1$, $K'_2$ and $K'_3$ are distinctly structure dependent constants
- $(\pi_i)_{123} =$ Internal pressure of the ternary mixture
In the present work, the equation (4) for ternary system is modified as

$$\log \pi_i = x_1 \log \pi_1 + x_2 \log \pi_2 + x_3 \log \pi_3 + x_1 x_2 (\beta_{12} (x_1 - x_2)) + x_2 x_3 (\beta_{23} (x_2 - x_3)) + x_3 x_1 (\beta_{31} (x_3 - x_1)) - C x_1 x_2 x_3$$  \hspace{1cm} (6)

Where

- $\beta_{12}$ = binary interaction constant for 1, 2 component
- $\beta_{23}$ = binary interaction constant for 2, 3 component
- $\beta_{31}$ = binary interaction constant for 3, 1 component

The constants $\beta_{12}$, $\beta_{23}$ and $\beta_{31}$ are determined from equation (4) using least square method. Constant C has been determined through a least square method at all the temperatures.

4. RESULTS AND DISCUSSION

For the Ternary and sub-binary systems, the internal pressure values are evaluated from the temperature range of 303.15K to 318.15K. These values are correlated through equations (4) and (6) is shown in Tables (2 & 3).

Table 2. Experimental and Calculated Internal pressures (in atm)

| Mol Frac | 303.15K | 308.15K | 313.15K | 318.15K |
|----------|---------|---------|---------|---------|
| x1       | EXP     | CAL     | EXP     | CAL     | EXP     | CAL     | EXP     | CAL     |
| 0.1016   | 2236.0  | 2225.2  | 2172.1  | 2166.3  | 2125.2  | 2115.0  | 2085.4  | 2073.9  |
| 0.201    | 2300.0  | 2289.4  | 2236.5  | 2227.3  | 2184.0  | 2174.1  | 2141.1  | 2130.5  |
| 0.301    | 2371.6  | 2363.7  | 2304.8  | 2298.5  | 2249.1  | 2243.2  | 2201.8  | 2197.0  |
| 0.4008   | 2449.0  | 2448.3  | 2377.8  | 2380.0  | 2322.2  | 2322.6  | 2274.4  | 2273.9  |
| 0.5017   | 2529.8  | 2545.3  | 2462.7  | 2474.1  | 2401.4  | 2414.5  | 2351.2  | 2363.1  |
| 0.6014   | 2645.2  | 2653.7  | 2575.0  | 2579.8  | 2511.5  | 2517.8  | 2456.5  | 2463.8  |
| 0.7033   | 2778.5  | 2778.6  | 2703.8  | 2702.1  | 2639.4  | 2637.7  | 2580.4  | 2580.9  |
| 0.7993   | 2914.4  | 2910.8  | 2834.5  | 2832.1  | 2764.4  | 2765.3  | 2705.0  | 2705.8  |
### System sec-butyl alcohol(1) + hexane(2)

| Mol Frac | 303.15K | 308.15K | 313.15K | 318.15K |
|----------|---------|---------|---------|---------|
| x1 EXP   | CAL     | EXP CAL | EXP CAL | EXP CAL |
| 0.1051   | 2251.821| 2242.171| 2197.044| 2186.876|
| 0.2041   | 2336.198| 2321.357| 2280.573| 2265.915|
| 0.3141   | 2432.489| 2421.634| 2377.999| 2365.713|
| 0.4023   | 2509.541| 2512.268| 2452.494| 2455.71 |
| 0.5058   | 2619.412| 2631.409| 2563.365| 2573.794|
| 0.5999   | 2741.206| 2752.911| 2680.968| 2694.021|
| 0.6989   | 2891.067| 2895.762| 2828.863| 2835.172|
| 0.8222   | 3099.669| 3097.818| 3034.703| 3034.54 |
| 0.9004   | 3251.553| 3241.483| 3188.756| 3176.129|

### System sec-butyl alcohol(1) + benzene(2)

| Mol Frac | 303.15K | 308.15K | 313.15K | 318.15K |
|----------|---------|---------|---------|---------|
| x1 EXP   | CAL     | EXP CAL | EXP CAL | EXP CAL |
| 0.1013   | 3276.863| 3263.685| 3203.142| 3190.139|
| 0.2018   | 3275.251| 3270.697| 3203.308| 3198.188|
| 0.3029   | 3286.307| 3282.14 | 3212.884| 3210.547|
| 0.4024   | 3294.1   | 3297.746| 3222.938| 3226.929|

β = 0.0715  β = 0.0759  β = 0.0777  β = 0.0801
ABSD = 0.2821 %  ABSD = 0.2136 %  ABSD = 0.2574 %  ABSD = 0.2676 %
Table 3. Experimental and Calculated Internal pressures (in atm) for the system benzene(1) + hexane(2) + sec-butyl alcohol(3)

| Mol Frac | 303.15K | 308.15K | 313.15K | 318.15K |
|----------|---------|---------|---------|---------|
| x1 | x2 | EXP | CAL | EXP | CAL | EXP | CAL | EXP | CAL |
| 0.1225 | 0.7894 | 2318.604 | 2372.167 | 2278.551 | 2317.373 | 2226.607 | 2267.348 | 2180.408 | 2226.415 |
| 0.2202 | 0.6873 | 2375.289 | 2440.105 | 2326.719 | 2387.119 | 2279.219 | 2337.243 | 2231.527 | 2295.946 |
| 0.3708 | 0.5247 | 2493.667 | 2548.28 | 2441.336 | 2496.091 | 2392.881 | 2445.825 | 2340.945 | 2403.063 |
| 0.4008 | 0.4967 | 2562.317 | 2571.26 | 2510.251 | 2518.581 | 2460.889 | 2467.873 | 2408.214 | 2424.626 |
| 0.4991 | 0.3992 | 2656.839 | 2674.647 | 2601.583 | 2620.553 | 2541.165 | 2568.28 | 2499.886 | 2523.352 |
| 0.6004 | 0.3002 | 2765.77 | 2814.948 | 2714.587 | 2758.523 | 2661.907 | 2703.879 | 2606.436 | 2656.798 |
| 0.6822 | 0.2354 | 2865.034 | 2927.488 | 2807.375 | 2867.366 | 2753.643 | 2809.668 | 2701.495 | 2760.357 |
| 0.7855 | 0.1322 | 3045.295 | 3086.881 | 2988.739 | 3021.499 | 2938.266 | 2960.151 | 2882.8 | 2907.386 |
| 0.1295 | 0.6904 | 2400.224 | 2440.035 | 2352.114 | 2386.885 | 2304.89 | 2338.778 | 2253.494 | 2296.96 |
| 0.2007 | 0.5985 | 2472.312 | 2481.023 | 2391.404 | 2431.291 | 2341.318 | 2384.726 | 2296.4 | 2343.179 |
| 0.2822 | 0.4966 | 2536.275 | 2533.49 | 2480.521 | 2486.263 | 2427.991 | 2440.84 | 2378.685 | 2399.095 |
| 0.4225 | 0.356 | 2644.701 | 2653.66 | 2596.796 | 2605.957 | 2546.943 | 2559.466 | 2496.079 | 2515.948 |
| 0.5212 | 0.2586 | 2771.755 | 2795.807 | 2718.175 | 2745.123 | 2670.034 | 2696.231 | 2609.653 | 2650.302 |
| C   | C   | C   | C   | ABSD % | ABSD % | ABSD % | ABSD % |
|-----|-----|-----|-----|--------|--------|--------|--------|
| 0.6061 | 0.1946 | 2882.3 | 2916.638 | 2832.512 | 2861.363 | 2786.356 | 2809.038 | 2731.935 | 2760.555 |
| 0.6766 | 0.1357 | 3039.526 | 3034.114 | 2987.92 | 2973.576 | 2926.137 | 2917.794 | 2879.654 | 2866.487 |
| 0.1205 | 0.5897 | 2513.491 | 2521.662 | 2458.413 | 2468.686 | 2411.655 | 2422.619 | 2365.829 | 2379.263 |
| 0.1999 | 0.4964 | 2569.775 | 2551.949 | 2528.281 | 2503.561 | 2476.672 | 2459.563 | 2441.955 | 2416.95 |
| 0.3215 | 0.3581 | 2669.675 | 2637.039 | 2614.556 | 2590.839 | 2557.943 | 2547.563 | 2509.907 | 2504.179 |
| 0.4218 | 0.257 | 2748.04 | 2770.923 | 2697.731 | 2722.382 | 2646.064 | 2677.179 | 2597.626 | 2631.461 |
| 0.4948 | 0.1917 | 2886.244 | 2887.913 | 2836.98 | 2835.369 | 2787.968 | 2787.437 | 2732.19 | 2739.264 |
| 0.5898 | 0.1203 | 3048.861 | 3040.701 | 2994.359 | 2980.958 | 3153.783 | 2928.345 | 2889.499 | 2876.473 |
| 0.1225 | 0.4927 | 2580.154 | 2606.145 | 2529.185 | 2552.861 | 2452.47 | 2508.629 | 2426.088 | 2463.65 |
| 0.2016 | 0.3763 | 2698.176 | 2673.83 | 2647.801 | 2624.081 | 2596.45 | 2581.822 | 2539.909 | 2536.684 |
| 0.3243 | 0.2543 | 2805.155 | 2792.252 | 2760.872 | 2742.343 | 2709.862 | 2699.014 | 2659.636 | 2652.11 |
| 0.4215 | 0.1764 | 2905.422 | 2915.624 | 2873.824 | 2861.956 | 2814.718 | 2815.688 | 2765.614 | 2766.188 |
| 0.4839 | 0.1156 | 3067.454 | 3044.866 | 3013.547 | 2985.109 | 2960.688 | 2935.52 | 2910.56 | 2882.718 |
| 0.1272 | 0.3973 | 2715.183 | 2715.181 | 2661.703 | 2660.105 | 2609.531 | 2616.79 | 2538.549 | 2569.456 |
| 0.2002 | 0.3186 | 2803.156 | 2763.374 | 2749.243 | 2711.049 | 2713.088 | 2668.554 | 2661.224 | 2621.245 |
| 0.2984 | 0.2205 | 2934.301 | 2869.548 | 2879.298 | 2816.384 | 2825.372 | 2772.81 | 3024.923 | 2723.775 |
| 0.3973 | 0.1128 | 3100.427 | 3069.24 | 3028.384 | 3007.933 | 2991.066 | 2960.133 | 2923.14 | 2906.1 |
| 0.1275 | 0.3 | 2893.766 | 2870.817 | 2808.343 | 2811.121 | 2756.968 | 2767.328 | 2695.534 | 2716.176 |
| 0.2 | 0.2187 | 2961.039 | 2943.526 | 2904.304 | 2884.664 | 2857.306 | 2841.071 | 2801.644 | 2789.106 |
| 0.3039 | 0.1155 | 3117.227 | 3103.552 | 3065.126 | 3039.731 | 3003.369 | 2993.429 | 2944.565 | 2937.968 |
| 0.1116 | 0.2182 | 3102.194 | 3036.133 | 3038.374 | 2969.591 | 2975.374 | 2924.819 | 2916.132 | 2869.075 |
| 0.1801 | 0.1396 | 3147.3 | 3126.911 | 3083.545 | 3059.496 | 3055.833 | 3014.475 | 3021.898 | 2957.275 |

| C = 1.0370 | C = 0.9539 | C = 0.9207 | C = 0.8938 |
|------------|------------|------------|------------|
| ABSD = 0.9816 % | ABSD = 1.0060 % | ABSD = 1.1969 % | ABSD = 1.3752 % |
Both the Experimental and Theoretical Internal pressure values are represented in
the tables (2 & 3). It is known from the tables that both the experimental and the theoretical
internal pressure values are more comparable with each other. By using least square
method, the Interaction constants (β) and (C) are evaluated from the temperature range of
303.15K to 318.15K. The absolute average deviation between the experimental and
correlated Internal Pressure values for all the binary systems varies from 0.17% to 0.38%.
The Interaction constant (β) evaluated for all the binary systems varies from 0.02 to 0.08.

It is known from the tables (2) & (3) that the increase in internal pressure values with
increase of alcohol concentration is probably due to Hydrogen bonding. Since alcohols are
strongly self-associated liquid having a three dimensional network of Hydrogen bond. So
it is apparent that when two interacting molecules are having some sort of attractive forces
like that of hydrogen bonding should result in increase of internal pressure. The hydrogen
bonding arises from short range interaction augmented by the fact that Hydrogen bond
distance (A-H--B) is greater than Vander Waals radii. The short range forces arise when the
molecules come close enough together causing a significant overlap of electron clouds and
are often highly directional.

The non-linear relationship is more obvious for sec-butyl alcohol and benzene. This
may be due to the interaction of the delocalized π-bond electron cloud with the sec-butyl
alcohol. Benzene is a non-polar and an inert aromatic solvent. The dissociation effect of
benzene molecule prevents self-association in associated alcohol molecules. The π-electron
cloud of benzene is responsible for the dissociation effect.

The Internal pressure varies with respect to the concentration and Temperature. This
is shown in graph. Figures 1-3 shows a linear variation of internal pressure with the
concentration for binary liquid mixtures from the temperature range of 303.15K to
318.15K.

![Graph showing the variation of internal pressure with concentration of benzene and hexane at various temperatures.](attachment:Fig_1.png)
From the Tables 2 & 3 it is known that the decrease in internal pressure values with increase of temperature is due to the decrease in cohesive forces. The reduction in internal pressure may be due to the loosening of cohesive forces leading to breaking the structure of the solute.
When the temperature is increased, there is a tendency for the solute molecules to move away from each other, reducing the possibility for the interaction, which may further reduce the cohesive forces. Due to weakening of intermolecular forces of attraction the internal pressure should fall. This is an important observation which we have made and which is to be expected theoretically also as cohesive forces between molecules becoming less with increasing temperature.

Figures (4-6) shows a linear variation of internal pressure with the reciprocal of temperature for binary liquid mixtures from the temperature range of 303.15K-313.15K. The absolute average deviation between the experimental and correlated values for the Ternary system varies from 0.98% to 1.37%.

The evaluated Interaction constant (C) for the Ternary system varies from 0.89 to 1.03.

![Fig. 4. Variation of Internal Pressure with Temperature of Benzene(1)+Hexane(2) system](image)

The average absolute deviation for the Ternary system is greater than that for the sub-binary system. This may be due to the Interaction between the three components. Among the three components, one of the component is self associated alcohol (sec-butyl alcohol). While the other two components (benzene and hexane) which are non-polar in nature.

Hexane is a non-polar chain molecule; only Vander Waals type interactions are present in hexane, while alcohol molecule is polar and associate strongly through hydrogen bonding. The alcohol molecule associate in inert hexane medium and form clusters. An associated molecular cluster in a liquid may be called as a quasi-molecule or a pseudo molecule. While with benzene, π-bond interaction takes place with alcohol.

Due to these strong interactions among the components in a ternary liquid mixture, the interaction constant values for the Ternary liquid mixture increases than that of the...
Binary liquid mixtures. This increase in Interaction constant values leads to increase in absolute average deviation for the Ternary system than that of the Binary system.

Fig. 5. **Variation of Internal Pressure with Temperature of sec-butyl alcohol(1)+Hexane(2) system**

![Graph for sec-butyl alcohol(1)+Hexane(2) system]

Fig. 6. **Variation of Internal Pressure with Temperature of sec-butyl alcohol(1)+Benzene(2) system**

![Graph for sec-butyl alcohol(1)+Benzene(2) system]
5. CONCLUSION

It is evident from the present work, that

- The absolute average deviation between the experimental and correlated values for the Binary system and Ternary system varies from 0.17% to 1.37% indicating the applicability of equations (4) and (5).
- Internal pressure and their use in the study of molecular interactions.
- The character of association of alcohols makes the study of interactions particularly interesting.
- The cohesive forces are of primary importance.

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