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# VISCOMETRIC STUDY ON BINARY LIQUID MIXTURES OF PROPIOPHENONE WITH ANILINE AND N-ALKYL SUBSTITUTED ANILINES, AT 303.15 TO 318.15 K

**Nanduri Gayatri Devi[1,✉], N.V.N.B. Srinivasa Rao[2], D. Ramachandran[3], V. Nagalakshmi[1], and P. Sunila Rani[4]**

[1]Department of Chemistry, Ch. S. D. St. Theresa's College (A) for Women, Eluru-534007, (Andhra Pradesh), India
[2]Department of Chemistry, DRG Government Degree College, Tadepallegudem, (AP) India
[3]Department of Chemistry, Acharya Nagarjuna University, Guntur, (AP) India
[4]Department of Microbiology, Ch. S. D. St. Theresa's College (A) for Women, Eluru-534007, (AP) India
✉Corresponding Author: gayatride081971@gmail.com

**ABSTRACT**

Densities and viscosities of binary mixtures of Propiophenone with Aniline, N-methylaniline, N, N- dimethylaniline, N, N- diethylaniline were measured over the entire composition range at $T$ = (303.15 to 318.15) K (with 5K interval) and atmospheric pressure. Experimental data were used to calculate the deviation of viscosity Δη, excess Gibb's free energy G*E activation of viscous flow for each binary system, and these excess thermodynamic properties were fitted to the Redlich-Kister polynomial equation to obtain the fitting coefficients and standard deviations. McAllister's three-body /four-body interaction models were used for the correlation of viscosity data. The studied systems exhibit good intermolecular interactions due to hydrogen ion transfer and charge dispersion in the carbonyl group and NH2 groups of Aniline and Alkyl Substituted Anilines. Experimental results are useful in various pharmaceutical industries.

**Keywords:** Viscosity, Deviation in Viscosity, McAllister's Models, Viscosity Relations.

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## INTRODUCTION

Viscosity properties of the binary liquid mixtures are important for fluid transport, chemical industries, pharmaceutical processes, and food products. Excess thermodynamic properties of the binary liquid mixtures of Propiophenone with Aniline, N-Methylaniline, N, N- Dimethyl aniline and N, N-Diethyl aniline were calculated based on the values of densities, ρ, and viscosities, η, over the entire composition range at $T$ = (303.15 to 318.15) K. Propiophenone is used as an intermediate in the industrial processes.[1] Aromatic anilines and substituted alkyl anilines are also useful in making dyes. Literature survey reveals some studies on Aniline with other compounds.[2–7] Hence, no studies were made on the current binary mixtures. Measured values of density and viscosity data were used to calculate Δη, one interaction parameters of d12, G*E, Wvis/RT, H12, and T12, (Grunberg and Nissan, Gibb's free energy, Katti and Chaudhary, Hind et.al., and Tamara-Kurata respectively) for the studied binary liquid mixtures. McAllister's three and four body, Auslander and Jouyban-Acree relations for two and three adjustable interaction parameter equations were used to correlate and to understand the intermolecular interactions. Experimental viscosity data was compared with the calculated values of having no adjustable parameter relations such as Bingham, Arrhenius, Kendall and Monroe, and Kendall. The values of Δη and G*E of binary mixtures were fitted to the Redlich Kister polynomial equation to estimate the coefficients and standard deviations between the experimental and theoretical values.

## EXPERIMENTAL

The purities of the selected solvents are as shown in Table-1 and they are purchased from S.D. Fine chemicals Ltd, India.


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Table -1: Provenance and Purity of the Materials Used

| Chemicals | CAS number | Source | Mass Fraction Purity |
|---|---|---|---|
| Propiophenone (PPH) | 93-55-0 | S.D fine Chemicals, India | 99.0% |
| Aniline (A) | 62-53-3 | S.D fine Chemicals, India | 99.7% |
| N-Methylaniline (MA) | 100-61-8 | S.D fine Chemicals, India | 99.5% |
| N,N-Diethylaniline (DMA) | 91-66-7 | S.D fine Chemicals, India | 99.6% |
| N,N-Dimethylaniline (DEA) | 121-69-7 | S.D fine Chemicals, India | 99.5% |

The water content was less than 0.003 mass%. Mettler Toledo (ME204) balance was used to weigh the pure liquids and prepare the liquid mixtures, precision of ± 0.1 mg. The experimental and literature values of densities and viscosities of the liquids are given in Table-2, a[8], b[9], c[10], d[11], e[11], f[7], g[12], h[7], i[1], j[13], k[14], l[2], m[15], n[16], o[17], p[18], q[19], r[20].

Table -2: Comparison of Experimental Density and Viscosity of Pure Liquid with Literature Values

| Compound | T(K) | Density (ρ) | | ref | Viscosity(η) | | ref |
|---|---|---|---|---|---|---|---|
| | | Experimental | Literature | | Experimental | Literature | |
| | | gm/cm$^3$ | gm/cm$^3$ | | mpa.s | mpa.s | |
| Propiophenone | 303.15 | 1.0045 | 1.0044 | j | 1.5150 | 1.5100 | h |
| | 308.15 | 1.0015 | 1.0060 | j | 1.4690 | 1.4690 | i |
| | 313.15 | 0.9985 | | | 1.4230 | | |
| | 318.15 | 0.9955 | | | 1.3770 | | |
| Aniline | 303.15 | 1.0128 | 1.0130 | a | 3.191 | 3.746 | n |
| | | | 1.0128 | b | | 3.770 | n |
| | | | 1.0128 | e | | 3.190 | l |
| | | | 1.0129 | l | | 3.190 | q |
| | | | 1.0132 | r | | | |
| | 308.15 | 1.0089 | 1.0087 | l | 2.811 | 2.800 | l |
| | 313.15 | 1.0049 | 1.0049 | e | 2.436 | 2.420 | l |
| | | | 1.0049 | k | | | |
| | 318.15 | 1.0009 | | | 2.065 | | |
| N-Methylaniline | 303.15 | 0.9783 | 0.9817 | c | 1.963 | 1.965 | c |
| | | | 0.9782 | e | | 1.963 | d |
| | | | 0.9783 | m | | | |
| | 308.15 | 0.9740 | 0.9742 | p | 1.811 | | |
| | 313.15 | 0.9696 | 0.9696 | e | 1.658 | | |
| | 318.15 | 0.9652 | | | 1.504 | | |
| N,N-DiMethylaniline | 303.15 | 0.9479 | 0.9480 | l | 1.173 | 1.174 | l |
| | 308.15 | 0.9436 | 0.9517 | f | 1.078 | 1.090 | l |
| | | | 0.9518 | g | | 1.111 | l |
| | | | 0.9436 | l | | | |
| | 313.15 | 0.9393 | 0.9394 | l | 0.981 | 0.982 | l |
| | 318.15 | 0.9351 | | | 0.885 | | |
| N,N-Diethylaniline | 303.15 | 0.9260 | 0.9260 | p | 1.703 | 1.703 | p |
| | | | 0.9253 | a | | 1.711 | a |
| | | | 0.9260 | o | | 1.703 | o |
| | 308.15 | 0.9217 | 0.9213 | a | 1.554 | 1.548 | a |
| | | | 0.9219 | o | | 1.550 | o |
| | 313.15 | 0.9176 | 0.9177 | p | 1.406 | 1.402 | p |
| | | | 0.9175 | o | | | |
| | 318.15 | 0.9134 | | | 1.263 | | |

**Apparatus and Procedures**

Anton Paar (DSA 5000 M) oscillating u-tube densimeter, automatically thermostatic within ±0.01K, was used to measure the densities of the pure components and the binary mixtures over the whole composition range *T*= (303.15 to 318.15) K. Averages of five measurements were taken at temperatures from 303.15 to 318.15K with an increment of 5K under atmospheric pressure. The standard uncertainties were found to be ±0.001 kg.m$^{-3}$ and ±0.005 mPa.s for the measurements of density and viscosity respectively. The Ubbelohde viscometer was used to measure the Viscosity as per the method described earlier.[21]

The measured values of densities[22], viscosities, and calculated values of deviation in viscosity and Gibbs free energy values with respective mole fractions of the studied binary mixtures at *T*=303.15 to 318.15K are tabulated in Table-3 and Table-4.

## RESULTS AND DISCUSSION

Negative values of deviation in viscosity (Δη) were observed and they were decreasing with the increase in temperature for all the studied binary mixtures. The difference in size and shape of the component molecules and the loss of dipolar association in pure components was the reason and it is in line with Fort and Moore (1966) and Pikkarainen (1983). The deviation in viscosity values are maximum at the mole fractions 0.5069, 0.4826, 0.4880 and 0.5447 are -0.0309, -0.0340, -0.0368 and -0.0416 for PPH+Aniline, PPH+MA, PPH+DMA and PPH+DEA at 303.15K, 313.15K and it is shown in the Figures-1-2. The order of interactions for deviation in viscosity is PPH+A>PPH+MA>PPH+DMA>PPH+DEA.

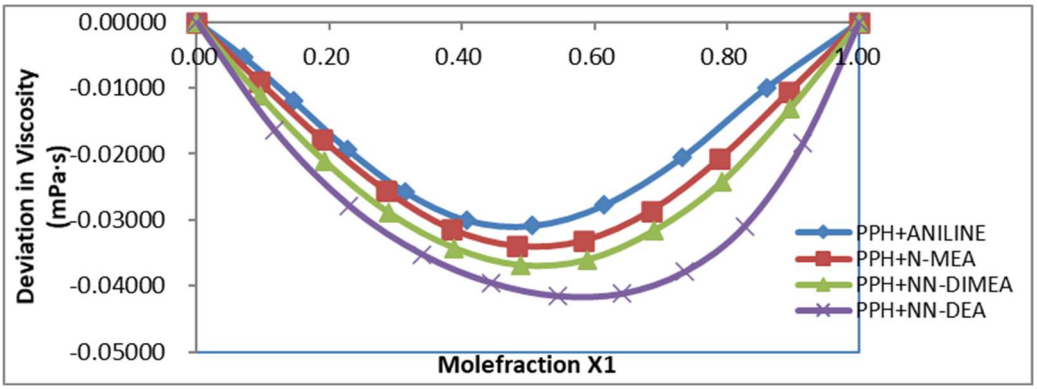


Fig.-1: Deviation in Viscosity Values of the Binary Mixtures Propiophenone with Aniline, N-Methylaniline, N, N-Dimethylaniline, N, N-Diethylaniline concerning their Mole Fraction Values at *T*= 303.15 K.

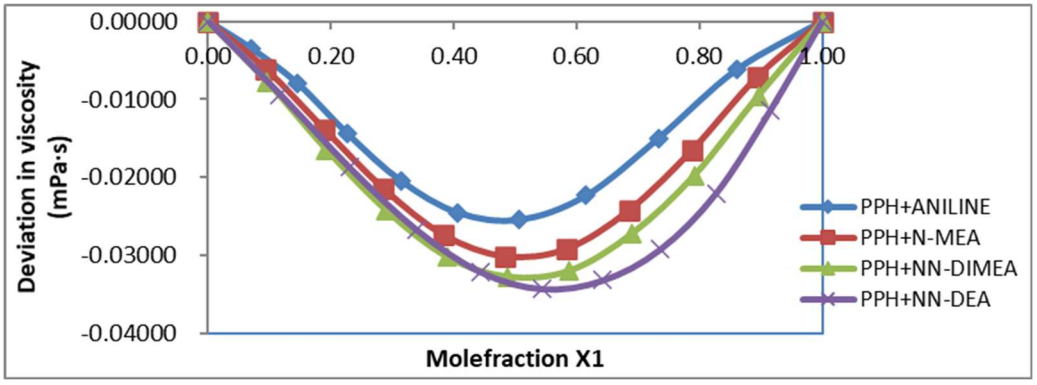


Fig.-2: Deviation in Viscosity Values of the Binary Mixtures Propiophenone with Aniline, N-Methylaniline, N, N-Dimethylaniline, N, N-Diethylaniline concerning their Mole Fraction Values at *T*= 313.15 K.

The presence of weak interactions between unlike molecules indicates a negative value of d12 and the presence of specific interactions indicates positive d12 values as per Moore et al[23] and Ramamoorthy.[24] Positive values of both deviations in viscosity, Δη, and Grunberg-Nissan parameter d12 indicates the

presence of strong specific interactions, negative Δη and positive d12 indicates weak specific interactions, negative values of both Δη and d12 indicates the absence of specific interactions as per Nigam and Mahal.[25]

Table -3 Density, Viscosity, Deviation in Viscosity and Gibbs Free Energy Values for the Binary Mixtures of Propiophenone with Aniline, N-Methylaniline, N, N-Dimethyl aniline & N, N-Diethyl aniline with respective Mole Fractions at T=(303.15- 308.15)K.

| X1 | 303.15K | | | | 308.15K | | | |
|---|---|---|---|---|---|---|---|---|
| | ρ | η | Δη | G*E | ρ | η | Δη | G*E |
| | gm/cm$^3$ | mPa·s | mPa·s | J/mol | gm/cm$^3$ | mPa·s | mPa·s | J/mol |
| PPH+ANILINE | | | | | | | | |
| 0.0000 | 1.0128 | 3.1911 | 0.0000 | 0.0000 | 1.0089 | 2.8107 | 0.0000 | 0.0000 |
| 0.0708 | 1.0122 | 3.0067 | -0.0053 | 0.4474 | 1.0083 | 2.6447 | -0.0041 | 0.3695 |
| 0.1463 | 1.0115 | 2.8279 | -0.0120 | 0.8470 | 1.0078 | 2.4844 | -0.0095 | 0.6934 |
| 0.2270 | 1.0109 | 2.6517 | -0.0193 | 1.1885 | 1.0072 | 2.3286 | -0.0158 | 0.9623 |
| 0.3136 | 1.0101 | 2.4794 | -0.0257 | 1.4669 | 1.0065 | 2.1832 | -0.0212 | 1.1772 |
| 0.4066 | 1.0093 | 2.3112 | -0.0300 | 1.6688 | 1.0059 | 2.0447 | -0.0252 | 1.3237 |
| 0.5069 | 1.0085 | 2.1458 | -0.0309 | 1.7765 | 1.0051 | 1.9131 | -0.0259 | 1.3969 |
| 0.6152 | 1.0076 | 1.9831 | -0.0277 | 1.7533 | 1.0043 | 1.7889 | -0.0232 | 1.3682 |
| 0.7327 | 1.0066 | 1.8259 | -0.0205 | 1.5388 | 1.0034 | 1.6743 | -0.0164 | 1.2009 |
| 0.8605 | 1.0056 | 1.6709 | -0.0099 | 1.0284 | 1.0025 | 1.5684 | -0.0072 | 0.8049 |
| 1.0000 | 1.0045 | 1.5150 | 0.0000 | 0.0000 | 1.0015 | 1.4690 | 0.0000 | 0.0000 |
| PPH+N-METHYL ANILINE | | | | | | | | |
| 0.0000 | 0.9783 | 1.9630 | 0.0000 | 0.0000 | 0.9740 | 1.8105 | 0.0000 | 0.0000 |
| 0.0939 | 0.9816 | 1.9036 | -0.0091 | -0.0157 | 0.9775 | 1.7530 | -0.0072 | -0.0251 |
| 0.1891 | 0.9848 | 1.8467 | -0.0179 | -0.0552 | 0.9809 | 1.7035 | -0.0160 | -0.0923 |
| 0.2856 | 0.9878 | 1.7930 | -0.0256 | -0.1061 | 0.9840 | 1.6604 | -0.0236 | -0.1619 |
| 0.3834 | 0.9907 | 1.7429 | -0.0314 | -0.1559 | 0.9870 | 1.6213 | -0.0294 | -0.2257 |
| 0.4826 | 0.9933 | 1.6967 | -0.0340 | -0.1857 | 0.9898 | 1.5864 | -0.0320 | -0.2616 |
| 0.5831 | 0.9958 | 1.6551 | -0.0332 | -0.1916 | 0.9924 | 1.5550 | -0.0312 | -0.2643 |
| 0.6852 | 0.9981 | 1.6175 | -0.0286 | -0.1646 | 0.9949 | 1.5274 | -0.0265 | -0.2216 |
| 0.7886 | 1.0003 | 1.5833 | -0.0207 | -0.1104 | 0.9972 | 1.5031 | -0.0186 | -0.1448 |
| 0.8935 | 1.0024 | 1.5501 | -0.0106 | -0.0455 | 0.9994 | 1.4844 | -0.0089 | -0.0572 |
| 1.0000 | 1.0045 | 1.5150 | 0.0000 | 0.0000 | 1.0015 | 1.4690 | 0.0000 | 0.0000 |
| PPH+N, N-DIMETHYLANILINE | | | | | | | | |
| 0.0000 | 0.9479 | 1.1730 | 0.0000 | 0.0000 | 0.9436 | 1.0778 | 0.0000 | 0.0000 |
| 0.0958 | 0.9538 | 1.1985 | -0.0112 | -0.1640 | 0.9498 | 1.1068 | -0.0089 | -0.0966 |
| 0.1925 | 0.9599 | 1.2243 | -0.0211 | -0.3095 | 0.9560 | 1.1370 | -0.0183 | -0.2203 |
| 0.2900 | 0.9659 | 1.2511 | -0.0289 | -0.4217 | 0.9621 | 1.1688 | -0.0261 | -0.3217 |
| 0.3886 | 0.9717 | 1.2802 | -0.0342 | -0.4912 | 0.9681 | 1.2030 | -0.0317 | -0.3918 |
| 0.4880 | 0.9774 | 1.3124 | -0.0368 | -0.5176 | 0.9739 | 1.2402 | -0.0343 | -0.4159 |
| 0.5885 | 0.9830 | 1.3475 | -0.0360 | -0.4920 | 0.9796 | 1.2803 | -0.0335 | -0.3917 |
| 0.6899 | 0.9884 | 1.3863 | -0.0316 | -0.4146 | 0.9851 | 1.3233 | -0.0291 | -0.3210 |
| 0.7922 | 0.9937 | 1.4278 | -0.0242 | -0.3042 | 0.9906 | 1.3692 | -0.0214 | -0.2164 |
| 0.8956 | 0.9990 | 1.4713 | -0.0131 | -0.1529 | 0.9960 | 1.4180 | -0.0109 | -0.0951 |
| 1.0000 | 1.0045 | 1.5150 | 0.0000 | 0.0000 | 1.0015 | 1.4690 | 0.0000 | 0.0000 |
| PPH+N,N-DIETHYLANILINE | | | | | | | | |
| 0.0000 | 0.9260 | 1.7030 | 0.0000 | 0.0000 | 0.9217 | 1.5544 | 0.0000 | 0.0000 |
| 0.1173 | 0.9340 | 1.6645 | -0.0165 | -0.1907 | 0.9300 | 1.5315 | -0.0129 | -0.1685 |
| 0.2302 | 0.9421 | 1.6318 | -0.0279 | -0.3321 | 0.9382 | 1.5114 | -0.0234 | -0.3152 |
| 0.3389 | 0.9502 | 1.6041 | -0.0352 | -0.4284 | 0.9464 | 1.4946 | -0.0308 | -0.4257 |
| 0.4437 | 0.9582 | 1.5800 | -0.0396 | -0.4932 | 0.9545 | 1.4807 | -0.0358 | -0.5040 |
| 0.5447 | 0.9661 | 1.5590 | -0.0416 | -0.5317 | 0.9626 | 1.4699 | -0.0380 | -0.5435 |
| 0.6422 | 0.9739 | 1.5411 | -0.0412 | -0.5406 | 0.9705 | 1.4624 | -0.0372 | -0.5402 |
| 0.7363 | 0.9816 | 1.5267 | -0.0379 | -0.5092 | 0.9783 | 1.4580 | -0.0335 | -0.4927 |
| 0.8272 | 0.9892 | 1.5166 | -0.0309 | -0.4252 | 0.9861 | 1.4574 | -0.0264 | -0.3904 |
| 0.9150 | 0.9968 | 1.5126 | -0.0183 | -0.2560 | 0.9938 | 1.4614 | -0.0148 | -0.2203 |

| X1 | 303.15K | | | | 308.15K | | | |
|---|---|---|---|---|---|---|---|---|
| | ρ | η | Δη | G*E | ρ | η | Δη | G*E |
| | gm/cm$^3$ | mPa·s | mPa·s | J/mol | gm/cm$^3$ | mPa·s | mPa·s | J/mol |
| 1.0000 | 1.0045 | 1.5150 | 0.0000 | 0.0000 | 1.0015 | 1.4690 | 0.0000 | 0.0000 |

Table -4 Density, Viscosity, Deviation in Viscosity and Gibbs Free Energy Values for the Binary Mixtures of Propiophenone with Aniline, N-Methylaniline, N, N-Dimethyl aniline & N, N-Diethyl aniline with respective Mole Fractions at T=(313.15- 318.15)K.

| X1 | 313.15K | | | | 318.15K | | | |
|---|---|---|---|---|---|---|---|---|
| | ρ | η | Δη | G*E | ρ | η | Δη | G*E |
| | gm/cm$^3$ | mPa·s | mPa·s | J/mol | gm/cm$^3$ | mPa·s | mPa·s | J/mol |
| PPH+ANILINE | | | | | | | | |
| 0.0000 | 1.0049 | 2.4360 | 0.0000 | 0.0000 | 1.0009 | 2.0650 | 0.0000 | 0.0000 |
| 0.0708 | 1.0044 | 2.2874 | -0.0035 | 0.2848 | 1.0005 | 1.9323 | -0.0027 | 0.2034 |
| 0.1463 | 1.0040 | 2.1460 | -0.0080 | 0.5292 | 1.0001 | 1.8071 | -0.0072 | 0.3592 |
| 0.2270 | 1.0035 | 2.0118 | -0.0144 | 0.7120 | 0.9997 | 1.6958 | -0.0136 | 0.4566 |
| 0.3136 | 1.0029 | 1.8920 | -0.0204 | 0.8449 | 0.9992 | 1.6011 | -0.0196 | 0.5155 |
| 0.4066 | 1.0023 | 1.7806 | -0.0245 | 0.9306 | 0.9987 | 1.5208 | -0.0237 | 0.5455 |
| 0.5069 | 1.0016 | 1.6833 | -0.0254 | 0.9676 | 0.9982 | 1.4540 | -0.0246 | 0.5539 |
| 0.6152 | 1.0009 | 1.5968 | -0.0223 | 0.9456 | 0.9976 | 1.4060 | -0.0215 | 0.5414 |
| 0.7327 | 1.0002 | 1.5251 | -0.0150 | 0.8404 | 0.9970 | 1.3756 | -0.0142 | 0.4968 |
| 0.8605 | 0.9994 | 1.4689 | -0.0062 | 0.5711 | 0.9963 | 1.3690 | -0.0054 | 0.3565 |
| 1.0000 | 0.9985 | 1.4230 | 0.0000 | 0.0000 | 0.9955 | 1.3770 | 0.0000 | 0.0000 |
| PPH+N-METHYL ANILINE | | | | | | | | |
| 0.0000 | 0.9696 | 1.6577 | 0.0000 | 0.0000 | 0.9652 | 1.5040 | 0.0000 | 0.0000 |
| 0.0939 | 0.9733 | 1.6049 | -0.0061 | -0.0386 | 0.9691 | 1.4573 | -0.0047 | -0.0429 |
| 0.1891 | 0.9768 | 1.5639 | -0.0138 | -0.1165 | 0.9727 | 1.4254 | -0.0123 | -0.1413 |
| 0.2856 | 0.9801 | 1.5304 | -0.0215 | -0.2070 | 0.9762 | 1.4026 | -0.0197 | -0.2471 |
| 0.3834 | 0.9832 | 1.5027 | -0.0274 | -0.2869 | 0.9794 | 1.3862 | -0.0259 | -0.3431 |
| 0.4826 | 0.9862 | 1.4788 | -0.0301 | -0.3300 | 0.9825 | 1.3727 | -0.0286 | -0.3894 |
| 0.5831 | 0.9889 | 1.4575 | -0.0292 | -0.3264 | 0.9855 | 1.3629 | -0.0276 | -0.3796 |
| 0.6852 | 0.9915 | 1.4401 | -0.0242 | -0.2671 | 0.9882 | 1.3557 | -0.0226 | -0.3073 |
| 0.7886 | 0.9940 | 1.4272 | -0.0165 | -0.1715 | 0.9908 | 1.3526 | -0.0149 | -0.1942 |
| 0.8935 | 0.9963 | 1.4210 | -0.0072 | -0.0614 | 0.9933 | 1.3591 | -0.0060 | -0.0670 |
| 1.0000 | 0.9985 | 1.4230 | 0.0000 | 0.0000 | 0.9955 | 1.3770 | 0.0000 | 0.0000 |
| PPH+N, N-DIMETHYLANILINE | | | | | | | | |
| 0.0000 | 0.9393 | 0.9807 | 0.0000 | 0.0000 | 0.9351 | 0.8850 | 0.0000 | 0.0000 |
| 0.0958 | 0.9459 | 1.0134 | -0.0077 | -0.0388 | 0.9419 | 0.9209 | -0.0062 | 0.0509 |
| 0.1925 | 0.9522 | 1.0481 | -0.0165 | -0.1248 | 0.9485 | 0.9600 | -0.0146 | 0.0074 |
| 0.2900 | 0.9584 | 1.0849 | -0.0243 | -0.2106 | 0.9548 | 1.0019 | -0.0223 | -0.0569 |
| 0.3886 | 0.9645 | 1.1243 | -0.0302 | -0.2787 | 0.9609 | 1.0463 | -0.0284 | -0.1152 |
| 0.4880 | 0.9704 | 1.1666 | -0.0328 | -0.2990 | 0.9670 | 1.0937 | -0.0312 | -0.1363 |
| 0.5885 | 0.9763 | 1.2117 | -0.0320 | -0.2784 | 0.9730 | 1.1438 | -0.0303 | -0.1218 |
| 0.6899 | 0.9820 | 1.2598 | -0.0273 | -0.2120 | 0.9788 | 1.1968 | -0.0257 | -0.0736 |
| 0.7922 | 0.9876 | 1.3107 | -0.0197 | -0.1300 | 0.9846 | 1.2525 | -0.0179 | -0.0143 |
| 0.8956 | 0.9931 | 1.3654 | -0.0096 | -0.0392 | 0.9902 | 1.3124 | -0.0083 | 0.0296 |
| 1.0000 | 0.9985 | 1.4230 | 0.0000 | 0.0000 | 0.9955 | 1.3770 | 0.0000 | 0.0000 |
| PPH+N,N-DIETHYLANILINE | | | | | | | | |
| 0.0000 | 0.9176 | 1.4064 | 0.0000 | 0.0000 | 0.9134 | 1.2626 | 0.0000 | 0.0000 |
| 0.1173 | 0.9260 | 1.3990 | -0.0094 | -0.1316 | 0.9220 | 1.2686 | -0.0074 | -0.1010 |
| 0.2302 | 0.9344 | 1.3915 | -0.0187 | -0.2745 | 0.9305 | 1.2722 | -0.0167 | -0.2517 |
| 0.3389 | 0.9427 | 1.3853 | -0.0267 | -0.4017 | 0.9389 | 1.2767 | -0.0247 | -0.3834 |
| 0.4437 | 0.9509 | 1.3816 | -0.0321 | -0.4915 | 0.9473 | 1.2832 | -0.0301 | -0.4748 |
| 0.5447 | 0.9591 | 1.3812 | -0.0343 | -0.5294 | 0.9555 | 1.2926 | -0.0323 | -0.5108 |
| 0.6422 | 0.9671 | 1.3839 | -0.0332 | -0.5148 | 0.9637 | 1.3049 | -0.0312 | -0.4925 |
| 0.7363 | 0.9751 | 1.3894 | -0.0292 | -0.4537 | 0.9718 | 1.3196 | -0.0272 | -0.4277 |
| 0.8272 | 0.9830 | 1.3981 | -0.0220 | -0.3415 | 0.9798 | 1.3372 | -0.0200 | -0.3115 |

| X1 | 313.15K | | | | 318.15K | | | |
|---|---|---|---|---|---|---|---|---|
| | ρ | η | Δη | G*E | ρ | η | Δη | G*E |
| | gm/cm$^3$ | mPa·s | mPa·s | J/mol | gm/cm$^3$ | mPa·s | mPa·s | J/mol |
| 0.9150 | 0.9908 | 1.4101 | -0.0115 | -0.1756 | 0.9877 | 1.3578 | -0.0095 | -0.1413 |
| 1.0000 | 0.9985 | 1.4230 | 0.0000 | 0.0000 | 0.9955 | 1.3770 | 0.0000 | 0.0000 |

Computed values of parameters (d12, Wvis/RT, H12, T12) are given in Table-5 at temperatures 303.15 K and 313.15 K for the binary mixtures of Propiophenone with Aniline, N-Methylaniline, N, N-Dimethylaniline and N, N-Diethylaniline.

Table-5: Grunberg - Nissan d12, Katti-chaudhari Wvis/RT, Hind et.al H12, and Tamara-Kurata T12 Constants for the Binary Mixtures of Propiophenone with Aniline, N-Methylaniline, N,N-Dimethylaniline and N,N-Diethylaniline computed by using Viscosity Data over the entire Composition Range at T=303.15K and 313.15K.

| X1 | $d_{12}$ | $W_{vis}/RT$ | $H_{12}$ | $T_{12}$ | $d_{12}$ | $W_{vis}/RT$ | $H_{12}$ | $T_{12}$ |
|---|---|---|---|---|---|---|---|---|
| | 303.15K | | | | 313.15K | | | |
| PPH+ANILINE | | | | | | | | |
| 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 |
| 0.0708 | 0.1993 | 0.2734 | 2.3125 | 2.4964 | 0.1018 | 0.1740 | 1.9029 | 2.0211 |
| 0.1463 | 0.1998 | 0.2725 | 2.3051 | 2.4684 | 0.0991 | 0.1703 | 1.8975 | 2.0020 |
| 0.2270 | 0.2010 | 0.2721 | 2.2981 | 2.4403 | 0.0931 | 0.1630 | 1.8885 | 1.9793 |
| 0.3136 | 0.2041 | 0.2738 | 2.2933 | 2.4141 | 0.0891 | 0.1577 | 1.8821 | 1.9588 |
| 0.4066 | 0.2098 | 0.2779 | 2.2909 | 2.3902 | 0.0878 | 0.1550 | 1.8787 | 1.9413 |
| 0.5069 | 0.2191 | 0.2855 | 2.2913 | 2.3696 | 0.0899 | 0.1555 | 1.8787 | 1.9275 |
| 0.6152 | 0.2328 | 0.2976 | 2.2945 | 2.3522 | 0.0966 | 0.1605 | 1.8824 | 1.9180 |
| 0.7327 | 0.2525 | 0.3157 | 2.3008 | 2.3385 | 0.1103 | 0.1724 | 1.8912 | 1.9143 |
| 0.8605 | 0.2827 | 0.3442 | 2.3119 | 2.3305 | 0.1309 | 0.1911 | 1.9037 | 1.9150 |
| 1.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 |
| PPH+N-METHYL ANILINE | | | | | | | | |
| 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 |
| 0.0939 | -0.0243 | -0.0074 | 1.6858 | 1.7163 | -0.0327 | -0.0182 | 1.5045 | 1.5232 |
| 0.1891 | -0.0306 | -0.0145 | 1.6807 | 1.7066 | -0.0449 | -0.0305 | 1.4952 | 1.5112 |
| 0.2856 | -0.0366 | -0.0209 | 1.6763 | 1.6977 | -0.0552 | -0.0408 | 1.4877 | 1.5008 |
| 0.3834 | -0.0420 | -0.0265 | 1.6726 | 1.6898 | -0.0631 | -0.0488 | 1.4824 | 1.4926 |
| 0.4826 | -0.0453 | -0.0299 | 1.6710 | 1.6842 | -0.0673 | -0.0531 | 1.4800 | 1.4876 |
| 0.5831 | -0.0470 | -0.0317 | 1.6708 | 1.6805 | -0.0680 | -0.0540 | 1.4803 | 1.4856 |
| 0.6852 | -0.0462 | -0.0307 | 1.6727 | 1.6793 | -0.0638 | -0.0497 | 1.4842 | 1.4876 |
| 0.7886 | -0.0425 | -0.0266 | 1.6768 | 1.6808 | -0.0555 | -0.0413 | 1.4909 | 1.4930 |
| 0.8935 | -0.0354 | -0.0192 | 1.6835 | 1.6853 | -0.0400 | -0.0259 | 1.5027 | 1.5036 |
| 1.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 |
| PPH+N, N-DIMETHYLANILINE | | | | | | | | |
| 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 |
| 0.0958 | -0.0731 | -0.0761 | 1.2791 | 1.2770 | -0.0111 | -0.0180 | 1.1572 | 1.1539 |
| 0.1925 | -0.0759 | -0.0800 | 1.2762 | 1.2741 | -0.0255 | -0.0323 | 1.1489 | 1.1459 |
| 0.2900 | -0.0779 | -0.0823 | 1.2737 | 1.2716 | -0.0348 | -0.0411 | 1.1430 | 1.1401 |
| 0.3886 | -0.0786 | -0.0831 | 1.2719 | 1.2699 | -0.0409 | -0.0471 | 1.1383 | 1.1357 |
| 0.4880 | -0.0788 | -0.0832 | 1.2703 | 1.2684 | -0.0422 | -0.0481 | 1.1362 | 1.1339 |
| 0.5885 | -0.0775 | -0.0816 | 1.2696 | 1.2679 | -0.0405 | -0.0462 | 1.1358 | 1.1337 |
| 0.6899 | -0.0744 | -0.0779 | 1.2702 | 1.2688 | -0.0343 | -0.0398 | 1.1382 | 1.1365 |
| 0.7922 | -0.0717 | -0.0742 | 1.2704 | 1.2694 | -0.0264 | -0.0317 | 1.1419 | 1.1408 |
| 0.8956 | -0.0644 | -0.0657 | 1.2740 | 1.2735 | -0.0118 | -0.0168 | 1.1507 | 1.1501 |
| 1.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 |
| PPH+N,N DIETHYLANILINE | | | | | | | | |
| 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 |
| 0.1173 | -0.0884 | -0.0740 | 1.5295 | 1.5219 | -0.0643 | -0.0511 | 1.3695 | 1.3722 |
| 0.2302 | -0.0891 | -0.0753 | 1.5302 | 1.5243 | -0.0753 | -0.0622 | 1.3619 | 1.3642 |
| 0.3389 | -0.0902 | -0.0768 | 1.5304 | 1.5259 | -0.0850 | -0.0720 | 1.3552 | 1.3573 |
| 0.4437 | -0.0934 | -0.0803 | 1.5288 | 1.5255 | -0.0930 | -0.0800 | 1.3497 | 1.3517 |

| X1 | $d_{12}$ | $W_{vis}$/RT | $H_{12}$ | $T_{12}$ | $d_{12}$ | $W_{vis}$/RT | $H_{12}$ | $T_{12}$ |
|---|---|---|---|---|---|---|---|---|
| | 303.15K | | | | 313.15K | | | |
| 0.5447 | -0.0993 | -0.0861 | 1.5252 | 1.5228 | -0.0988 | -0.0858 | 1.3456 | 1.3476 |
| 0.6422 | -0.1080 | -0.0945 | 1.5193 | 1.5179 | -0.1031 | -0.0900 | 1.3425 | 1.3443 |
| 0.7363 | -0.1193 | -0.1054 | 1.5114 | 1.5107 | -0.1071 | -0.0939 | 1.3395 | 1.3411 |
| 0.8272 | -0.1341 | -0.1195 | 1.5009 | 1.5007 | -0.1093 | -0.0960 | 1.3376 | 1.3388 |
| 0.9150 | -0.1479 | -0.1323 | 1.4911 | 1.4912 | -0.1042 | -0.0907 | 1.3409 | 1.3415 |
| 1.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 | 0.0000 |

Table-6: The Standard Deviation Percentages (%) for the Binary Systems at *T*=303.15K to 318.15K calculated from the Kendall-Monroe, Bingham, Arrhenius-Eyring's, Kendall Equations.

| T/K | Kendall-Munroe | Bingham | Arrhenius | Kendall | Kendall-Munroe | Bingham | Arrhenius | Kendall |
|---|---|---|---|---|---|---|---|---|
| | σ (%) | σ (%) | σ (%) | σ (%) | σ (%) | σ (%) | σ (%) | σ (%) |
| | | PPH+ANILINE | | | | PPH+N-MEA | | |
| 303.15k | 2.22 | 0.85 | 8.15 | 3.74 | 0.94 | 1.33 | 0.17 | 0.74 |
| 308.15K | 1.59 | 0.77 | 6.65 | 2.75 | 1.05 | 1.31 | 0.27 | 0.92 |
| 313.15K | 0.83 | 0.82 | 4.92 | 1.64 | 1.14 | 1.28 | 0.60 | 1.07 |
| 318.15K | 0.17 | 0.88 | 3.06 | 0.56 | 1.23 | 1.27 | 0.94 | 1.21 |
| | | PPH+NN-DMA | | | | PPH+NN-DEA | | |
| 303.15k | 1.54 | 1.92 | 1.57 | 1.34 | 1.85 | 1.93 | 2.24 | 1.81 |
| 308.15K | 1.29 | 1.85 | 1.26 | 1.01 | 1.79 | 1.81 | 2.00 | 1.78 |
| 313.15K | 1.04 | 1.85 | 0.92 | 0.64 | 1.67 | 1.67 | 1.67 | 1.67 |
| 318.15K | 0.71 | 1.84 | 0.47 | 0.22 | 1.61 | 1.66 | 1.33 | 1.59 |

Under the current study, the binary mixture of PPH+Aniline shows positive values of d12 and fewer negative values of Δη, it indicates weak specific interactions whereas the other three binary mixtures are having negative values of both Δη and d12 which indicates the absence of specific interactions. Hence, The PPH+Aniline have weak specific interactions, while the other three have no specific interactions, therefore the structural effect is dominant. Under the current study, positive values of G*E for the binary mixture PPH+Aniline indicate dipole-dipole interactions between the component molecules. The negative values of G*E for the remaining three binary mixtures show the dispersion forces and it is supported by Reed and Taylor.[26] The negative values of the single adjustable parameter of Katti-Chaudhary (Wvis/RT) suggest weak interactions and positive values for strong interactions. The values Wvis/RT of binary liquid mixture PPH+Aniline are less positive for the entire range of composition, whereas values of Wvis/RT for the other three binary liquid mixtures PPH+MA, DMA and DEA are negative.

The single interaction parameter values of Tamara-Kurata, T12, and Hind et.al, H12, don't change considerably from each other. This is in agreement with the Fort and Moore[23]. Positive values are observed for all the four binary liquid mixtures over the entire composition range and the values are decreasing with an increase in temperatures from 303.15 to 318.15K. Hence, it shows weak specific interactions.

Average standard deviation percentage (σ%) values were calculated and tabulated in Table-5. Amongst all, mixed results were observed. Standard deviation percentage values of Bingham viscosity relation in PPH+A binary mixture, Arrhenius-Eyring's values in PPH+N-MEA binary mixture and Kendall values in PPH+NN-DMA and PPH+NN-DEA binary mixtures are in good agreement with the experimental data, as their σ% values are nearest to zero. McAllister's three/four body equations, Auslander and Jouyban–Acree relations were used to correlate the viscosity data. Two and three adjustable parameters and the standard deviations were calculated with the method of least squares. Interaction coefficients from these relations and standard deviation values are tabulated in Table-7 and Table-8.

Comparatively, obtained standard deviation values are much better with McAllister's four-body model and Jouban-Acree relations and are having a good agreement with the experimental data with the three adjustable interaction parameters. These parameters are very much useful for the chemical and industrial process and design of equipment. With this study, an increase in adjustable parameters reduces the standard deviation and enhances the correlating ability for the binary liquid mixtures.

Table-7:Calculated Values of Interaction parameters of McAllister three body and Four Body Equations for the Binary Mixtures of Propiophenone with Aniline, N-Methylaniline, N, N-Dimethylaniline and N, N-Diethylaniline.

| Binary Mixture | T/K | McAllister Three Body | | | McAllister Four Body | | | |
|---|---|---|---|---|---|---|---|---|
| | | a | b | σ | A | B | C | σ |
| PPH+ANILINE | 303.15 | 2.1503 | 2.6499 | 0.0019 | 1.9815 | 2.3434 | 2.8042 | 0.0002 |
| | 308.15 | 1.962 | 2.3726 | 0.0023 | 1.8386 | 2.1237 | 2.5022 | 0.0002 |
| | 313.15 | 1.7823 | 2.0974 | 0.0025 | 1.699 | 1.8978 | 2.205 | 0.0002 |
| | 318.15 | 1.6107 | 1.8267 | 0.0024 | 1.5627 | 1.6775 | 1.9094 | 0.0001 |
| PPH+N-MEA | 303.15 | 1.6273 | 1.7869 | 0.0009 | 1.6065 | 1.6877 | 1.8381 | 0.0001 |
| | 308.15 | 1.5547 | 1.6631 | 0.0018 | 1.5387 | 1.5836 | 1.7161 | 0.0001 |
| | 313.15 | 1.4671 | 1.5527 | 0.0018 | 1.471 | 1.4803 | 1.5938 | 0.0002 |
| | 318.15 | 1.3867 | 1.4353 | 0.0022 | 1.4021 | 1.3775 | 1.47 | 0.0001 |
| PPH+NN-DMA | 303.15 | 1.3589 | 1.2442 | 0.0005 | 1.4007 | 1.2928 | 1.2297 | 0.0001 |
| | 308.15 | 1.3017 | 1.1721 | 0.0012 | 1.3514 | 1.2192 | 1.1559 | 0.0001 |
| | 313.15 | 1.2432 | 1.0975 | 0.0017 | 1.2989 | 1.1471 | 1.0777 | 0.0001 |
| | 318.15 | 1.1848 | 1.0254 | 0.0023 | 1.2471 | 1.0756 | 1.0016 | 0.0002 |
| PPH+NN-DEA | 303.15 | 1.5209 | 1.6135 | 0.0054 | 1.498 | 1.574 | 1.6146 | 0.0002 |
| | 308.15 | 1.4422 | 1.4986 | 0.0034 | 1.4397 | 1.4674 | 1.504 | 0.0001 |
| | 313.15 | 1.3643 | 1.3849 | 0.0014 | 1.3815 | 1.3624 | 1.3937 | 0.0002 |
| | 318.15 | 1.2906 | 1.2753 | 0.0017 | 1.3185 | 1.2641 | 1.2804 | 0.0003 |

Table-8**:** Calculated Values of Interaction Parameters of Auslander and Jouyban-Acree equations for the Binary Mixtures of Propiophenone with Aniline, N-Methylaniline, N, N-Dimethylaniline and N, N-Diethylaniline.

| Binary Mixture | T/K | Auslander Equation | | | | Jouyban-Acree Equation | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | B12 | A21 | B21 | σ | Ao | A1 | A2 | σ |
| PPH+ANILINE | 303.15 | 1.0157 | 0.9482 | 0.9854 | 0.0026 | 66.1868 | 16.8143 | 0.7687 | 0.0002 |
| | 308.15 | 1.0175 | 0.9471 | 0.9853 | 0.0027 | 48.6729 | 11.5753 | 1.0882 | 0.0002 |
| | 313.15 | 1.022 | 0.9356 | 0.9778 | 0.0033 | 28.1379 | 6.6402 | 2.2588 | 0.0002 |
| | 318.15 | 0.0707 | 0.0696 | 12.5029 | 0.0031 | 7.5925 | 3.0158 | 5.8035 | 0.0001 |
| PPH+N-MEA | 303.15 | 0.3648 | 0.299 | 2.3848 | 0.0011 | -13.8701 | -2.7334 | -2.7884 | 0.0001 |
| | 308.15 | 0.1779 | 0.1543 | 4.2745 | 0.0013 | -17.7313 | -2.4692 | -4.581 | 0.0001 |
| | 313.15 | 0.102 | 0.101 | 5.5888 | 0.0016 | -21.2056 | -2.0448 | -7.5051 | 0.0002 |
| | 318.15 | 0.135 | 0.14 | 2.1938 | 0.0016 | -24.3984 | -1.6514 | -11.6921 | 0.0001 |
| PPH+NN-DMA | 303.15 | 0.8609 | 1.4279 | 1.0046 | 0.0006 | -23.7882 | -0.0562 | -73.3776 | 0.0004 |
| | 308.15 | 4.8666 | 7.0244 | 0.1798 | 0.0011 | -19.0828 | -0.1516 | -66.5628 | 0.0003 |
| | 313.15 | 1.1144 | 1.5136 | 0.8517 | 0.0021 | -13.2524 | -0.1754 | -83.4016 | 0.0002 |
| | 318.15 | 1.1806 | 1.5136 | 0.8237 | 0.0027 | -4.9297 | 0.0324 | 627.197 | 0.0004 |
| PPH+NN-DEA | 303.15 | 6.0009 | 3.2874 | -0.1735 | 0.0005 | -29.2444 | -10.0341 | 1.0303 | 0.0002 |
| | 308.15 | 4.2828 | 1.7901 | -0.8049 | 0.0002 | -30.3602 | -9.5487 | 0.2949 | 0.0001 |
| | 313.15 | -8.0608 | 1.9897 | 4.8771 | 0.0003 | -30.2132 | -9.1968 | -0.6052 | 0.0002 |
| | 318.15 | 3.5332 | 13.5872 | 0.1175 | 0.0011 | -29.9469 | -8.9747 | -1.2021 | 0.0003 |

The values of deviations in viscosity (Δη) and excess Gibbs Energy (G*E) concerning the mole fraction at temperatures from 303.15K to 318.15 K were fitted to the Redlich–Kister Equation[27]of the type are tabulated in Table-9.

Table -9: Coefficients of Redlich-Kister Polynomial Equation and Standard **D**eviation of **B**inary **S**ystems.

| Property | Temp (K) | A0 | A1 | A2 | A3 | A4 | σ | A0 | A1 | A2 | A3 | A4 | σ |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | PPH+ANILINE | | | | | | PPH+N-METHYL ANILINE | | | | | |
| Δη (mPa.s) | 303.15 | -0.1238 | 0.0137 | 0.0654 | -0.0096 | 0.0033 | 0.0001 | -0.1365 | -0.0074 | 0.0460 | 0.0084 | -0.0037 | 0.0000 |
| | 308.15 | -0.1043 | 0.0117 | 0.0699 | -0.0016 | -0.0006 | 0.0001 | -0.1286 | -0.0056 | 0.0548 | 0.0046 | 0.0094 | 0.0001 |
| | 313.15 | -0.1023 | 0.0148 | 0.0978 | -0.0108 | -0.0255 | 0.0002 | -0.1211 | -0.0063 | 0.0747 | 0.0085 | -0.0004 | 0.0001 |

| Property | Temp (K) | A0 | A1 | A2 | A3 | A4 | σ | A0 | A1 | A2 | A3 | A4 | σ |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | 318.15 | -0.0990 | 0.0151 | 0.0978 | -0.0120 | -0.0119 | 0.0001 | -0.1149 | -0.0053 | 0.0838 | 0.0046 | 0.0042 | 0.0001 |
| $G^{*E}$ (J/mol) | 303.15 | 7.0901 | 1.0985 | 1.0537 | 0.2667 | 0.1203 | 0.0009 | -0.7584 | -0.2921 | 0.6914 | 0.1932 | -0.0328 | 0.0006 |
| | 308.15 | 5.5759 | 0.6412 | 1.0269 | 0.3030 | 0.0694 | 0.0009 | -1.0615 | -0.2614 | 0.8621 | 0.1753 | 0.1423 | 0.0016 |
| | 313.15 | 3.8616 | 0.2794 | 1.3568 | 0.1672 | -0.2768 | 0.0018 | -1.3336 | -0.2521 | 1.2408 | 0.2468 | -0.0224 | 0.0010 |
| | 318.15 | 2.2112 | 0.0123 | 1.4521 | 0.0998 | -0.1562 | 0.0016 | -1.5690 | -0.2009 | 1.4756 | 0.1769 | 0.0376 | 0.0018 |
| | | PPH+NN-DIETHYLANILINE | | | | | | PPH+NN-DIMETHYLANILINE | | | | | |
| Δη (mPa.s) | 303.15 | -0.1637 | -0.0375 | -0.0607 | -0.0140 | 0.0160 | 0.0001 | -0.1474 | -0.0102 | 0.0164 | 0.0058 | 0.0042 | 0.0001 |
| | 308.15 | -0.1490 | -0.0408 | -0.0232 | -0.0028 | 0.0172 | 0.0001 | -0.1374 | -0.0107 | 0.0329 | 0.0044 | 0.0160 | 0.0000 |
| | 313.15 | -0.1345 | -0.0432 | 0.0142 | 0.0045 | 0.0162 | 0.0002 | -0.1317 | -0.0110 | 0.0509 | 0.0051 | 0.0071 | 0.0001 |
| | 318.15 | -0.1263 | -0.0436 | 0.0157 | 0.0062 | 0.0477 | 0.0002 | -0.1251 | -0.0121 | 0.0628 | 0.0046 | 0.0114 | 0.0001 |
| $G^{*E}$ (J/mol) | 303.15 | -2.0682 | -0.7376 | -0.9036 | -0.2330 | 0.2601 | 0.0015 | -2.0636 | 0.1271 | 0.4005 | 0.0556 | 0.0939 | 0.0020 |
| | 308.15 | -2.1178 | -0.7576 | -0.3429 | -0.0269 | 0.2994 | 0.0017 | -1.6601 | 0.1236 | 0.7365 | -0.0541 | 0.2863 | 0.0012 |
| | 313.15 | -2.0691 | -0.7584 | 0.3070 | 0.0936 | 0.2748 | 0.0032 | -1.1975 | 0.0997 | 1.0999 | -0.1101 | 0.1270 | 0.0023 |
| | 318.15 | -1.9984 | -0.7323 | 0.4176 | 0.0925 | 0.7941 | 0.0046 | -0.5470 | 0.0206 | 1.3803 | -0.2472 | 0.2620 | 0.0013 |

## CONCLUSION

Densities and viscosities values were measured for the studied binary liquid mixtures at temperatures, $T$ = (303.15 to 318.15) K. The G*E, Wvis/RT, H12, and d12 values were positive for Propiopheone + Aniline mixtures over the experimental temperature range, while they were negative for Propiophenone+N-Methylaniline, Propiophenone+N, N-Dimethylaniline and Propiophenone+N, N-Diethylaniline. The positive G*E, Wvis/RT, H12, and d12 values were attributed to the formation of the new complex, whereas negative values were due to the consequence of dominating effect. The calculated values of Δη and G*E were correlated with the Redlich-Kister polynomial equation to compute the coefficients and standard deviations. Viscosity data compared with several viscosity relations like Kendall-Monroe, Bingham, Arrhenius, Kendall and calculated the two / three adjustable interaction parameters and respective standard deviations by correlating with the McAllister three/four body models, Jouban-Acree and Auslander equations. The obtained results are useful in various industrial and chemical processes.

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