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Ultrasonic study of hydrogen bonded complexes in benzene solution

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ABSTRACT

Ultrasonic Velocity and density measurements are reported for H- Bonded complexes of n-Butylamine + o-chlorophenol, n-Butylamine + p-chlorophenol, n-Propylamine + o-chlorophenol, n-Propylamine + p-chlorophenol, Trimethylamine + m-chlorophenol, Aniline + m-chlorophenol in benzene solution, at 2MHZ and at 313K. Various ultrasonic parameters, such as adiabatic compressibility (βa), Rao's constant (R), Free Length (L_f), and available volume (Va) are reported. Using these parameters the strength of H- Bonded complex is compared.

Keywords: Ultrasonic velocity, adiabatic compressibility, Rao's constant, Free Length, n-Butylamine, n-Propylamine, Trimethylamine, Aniline, o-chlorophenol, p-chlorophenol, m-chlorophenol and H- Bonded complex.

INTRODUCTION

The complex formation in the liquid mixtures can be detected by maximum or minimum or any abrupt change from the normal behavior in the physical properties [1-5]. Many attempts [6-15] have been made to study ultrasonic velocities of binary liquid mixtures and relate them to find other parameters. Nambinarayanan et al [16] measured ultrasonic velocity and density in oxalic acid, benzoic acid and succinic acid in dioxane at the temperature range 303K - 333K. The result showed molecular association through hydrogen binding. Rao et al [17] reported "Clathrate like" structure of water and alcohol. Sosamma et al [18] reported complex formation between o and p-hydroxy benzoic acid in dioxane at 308K, 313K and 323K respectively by measuring ultrasonic velocity at various solute concentrations. Orzechowski [19] reported ultrasonic velocity and density of solution of phenol, k-naphthol and benzoic acid in benzene and dioxane.

Keeping this in view, in the present paper, ultrasonic velocity (U) and density (ρ) measurements are reported in the following H- bonded complexes in benzene solution.

1. n-Butylamine (nBA) + o-chlorophenol (oCLP),
2. n-Butylamine (nBA) + p-chlorophenol (pCLP),
3. n-Propylamine (nPA) + o-chlorophenol (oCLP),
4. n-Propylamine (nPA) + p-chlorophenol (pCLP),
5. Trimethylamine(TEA) + m-chlorophenol(mCLP) and
6. Aniline (ANL) + m-chlorophenol (mCLP).

MATERIALS AND METHODS

The liquids were used of BDR AR grade and were redistilled before use. Low weight fraction mixtures of above constituents were made using micropipettes' having accuracy of 0.02ml. These mixtures were dispersed in 25ml benzene before measurements. Ultrasonic velocity measurements were made using ultrasonic interferometer (Mittal, Model F-83) at 2MHz Frequency. The accuracy of Velocity measurement was ±3 m/s. Density measurements were made using standard density bottle (10 ml, Corning make). Monopan balance of accuracy of 0.01mg was used for weight measurements. All the measurements were made at 313K temperature.

Various acoustical and thermodynamical parameters were calculated from the measured data using the formulations, are as follows:

Molar Volume (V_M) = $M / \rho_{ABS} = ((M_A X_A + M_B X_B + M_S X_S) / \rho_{ABS})$ --------------- (1)

Adiabatic Compressibility (βa) = $1 / (U^2 \rho_{ABS})$ --------------- (2)

Available Volume (Va) = $V_M (1-(U/U_\infty))$ --------------- (3)

Free Length (L_f) = $(K / (U_* \rho_{ABS}^{1/2}))$ --------------- (4)

Rao's constant (R) = $U^{1/3} {}_* V_M$ --------------- (5)

Where U is the Ultrasonic Velocity, M is the molecular weight, x – weight fraction, ρ_{ABS}- density of the mixture, U_∞= 1600m/s and K is temperature dependent Jacobson's constant.

RESULTS AND DISCUSSION

The graphs of ultrasonic velocity (U) and adiabatic compressibility (βa) are plotted vs. weight fraction of amines (A) (Fig.-1 to 6). Molar Volume (V_M), Available Volume (Va), Free Length (L_f) and Rao's constant (R) respectively are evaluated and reported in Tables-1 to 6. Maximum, observed in ultrasonic velocity vs. concentration curve for n-Butylamine (nBA) + p-chlorophenol (pCLP) in benzene, indicates complex, 10 & 11 formation of O-H…N type near 1:1 concentration. This is again supported by minimum in compressibility, Free Length and available volume at this concentration. On the other hand ultrasonic velocity curve for n-Butylamine (nBA) + o-chlorophenol (oCLP) in benzene has slightly convex upward trend. Chlorine in the ortho-position of oCLP reduces the volume of closest approach with nBA and as such the molecular interaction is reduced between n-Butylamine (nBA) + o-chlorophenol (oCLP) than n-Butylamine (nBA) + p-chlorophenol (pCLP). This gets further support as free length and available volume for n-Butylamine (nBA) + o-chlorophenol (oCLP) are concentration at which velocity peak occurs.

Similar behavior is observed in n-Propylamine (nPA) + p-chlorophenol (pCLP) and n-Propylamine (nPA) + o-chlorophenol (oCLP) in benzene. In n-Propylamine (nPA) + o-chlorophenol (oCLP), O-H...N interactions are reduces due to steric nature of oCLP. If comparison is made in n-Propylamine (nPA) + p-chlorophenol (pCLP) and n-Butylamine (nBA) + p-chlorophenol (pCLP), it appears from ultrasonic velocity curves that pCLP interacts more with nBA than nPA. This is also confirmed from lower values of free length and available volume for n-Butylamine (nBA) + p-chlorophenol (pCLP) than n-Propylamine (nPA) + p-chlorophenol (pCLP) at the peak of ultrasonic velocity. This may be due to stronger basic nature of nBA.

Trimethylamine (TEA) + m-chlorophenol (mCLP) in benzene has convex upward trend while Aniline (ANL) + m-chlorophenol (mCLP) has concave upward trends. This indicates that extend of interaction of mCLP is more with Trimethylamine (TEA) than Aniline (ANL). This may be due to more basicity of Trimethylamine (TEA) than Aniline (ANL).

1. Tables: Molar Volume (V_M), Available Volume (Va), Free Length (L_f) and Rao's constant (R) respectively are evaluated and reported in Tables-1 to 6.

Table-1: nBA (A) + oCLP (B) in Benzene(S)

X_A	V_M (cm^2mole^{-1})	$\beta a \times 10^{-11}$ (cm^2dyne^{-1})	$L_f \times 10^{-10}$ (m)	Va (cm^3mole^{-1})	R
0.0000	92.98	7.25	0.5466	20.92	4637.00
0.0127	92.78	7.31	0.5490	20.77	4628.00
0.0192	93.38	7.44	0.5537	21.04	4655.00
0.0258	91.68	7.28	0.5480	20.34	4577.00
0.0323	91.32	7.42	0.5531	20.80	4548.00
0.0390	91.80	7.44	0.5539	20.59	4578.00
0.0456	91.27	7.47	0.5548	20.52	4551.00
0.0592	91.03	7.56	0.5586	20.49	4539.00
0.0661	90.73	7.72	0.5640	20.85	4569.00

Table-2: nBA (A) + pCLP (B) in Benzene (S)

X_A	V_M (cm^2mole^{-1})	$\beta a \times 10^{-11}$ (cm^2dyne^{-1})	$L_f \times 10^{-10}$ (m)	Va (cm^3mole^{-1})	R
0.0000	93.92	7.32	0.5490	20.09	4684.00
0.0127	94.41	7.40	0.5522	20.95	4714.00
0.0192	93.13	7.39	0.5520	20.89	4645.00
0.0257	92.69	7.39	0.5520	20.72	4624.00
0.0323	92.43	7.27	0.5472	19.87	4628.00
0.0389	92.16	7.39	0.5521	20.31	4604.00
0.0456	92.15	7.45	0.5540	20.29	4604.00
0.0524	91.69	7.50	0.5559	20.36	4577.00
0.0592	91.48	7.59	0.5592	20.55	4562.00
0.0661	91.50	7.70	0.5633	20.80	4558.00

Table-3: nPA (A) + oCLP (B) in Benzene (S)

X_A	V_M (cm^2mole^{-1})	$\beta a \times 10^{-11}$ (cm^2dyne^{-1})	$L_f \times 10^{-10}$ (m)	Va (cm^3mole^{-1})	R
0.0000	93.25	7.27	0.5475	20.95	4650.00
0.0116	93.23	7.36	0.5509	20.86	4651.00
0.0175	92.91	7.39	0.5517	20.80	4635.00
0.0234	92.25	7.37	0.5512	20.51	4605.00
0.0295	91.98	7.46	0.5547	19.72	4586.00
0.0355	91.51	7.49	0.5557	20.46	4561.00
0.0416	91.44	7.53	0.5570	20.54	4560.00
0.0478	91.07	7.57	0.5587	20.51	4540.00
0.0540	90.62	7.58	0.5590	20.33	4520.00
0.0603	90.44	7.66	0.5621	20.45	4507.00

Table-4: nPA (A) + pCLP (B) in Benzene (S)

X_A	V_M (cm^2mole^{-1})	$\beta a \times 10^{-11}$ (cm^2dyne^{-1})	$L_f \times 10^{-10}$ (m)	Va (cm^3mole^{-1})	R
0.0000	93.42	7.28	0.5475	20.98	4659.00
0.0174	93.37	7.36	0.5507	20.56	4665.00
0.0234	92.92	7.35	0.5502	21.30	4645.00
0.0294	92.35	7.34	0.5499	20.07	4620.00
0.0354	91.95	7.44	0.5537	20.34	4592.00
0.0416	91.36	7.43	0.5535	20.12	4564.00
0.0478	91.34	7.63	0.5607	20.74	4550.00
0.0540	90.94	7.61	0.5599	20.40	4535.00
0.0603	90.44	7.67	0.5621	20.45	4507.00

Table-5: TEA (A) + mCLP (B) in Benzene (S)

X_A	V_M (cm^2mole^{-1})	$\beta a \times 10^{-11}$ (cm^2dyne^{-1})	$L_f \times 10^{-10}$ (m)	Va (cm^3mole^{-1})	R
0.0000	92.73	7.93	0.5717	23.72	4561.00
0.0050	92.70	7.74	0.5647	22.79	4581.00
0.0150	93.11	7.96	0.5727	23.43	4589.00
0.0307	92.67	8.29	0.5845	24.48	4541.00
0.0360	92.64	8.34	0.5863	24.54	4538.00
0.0413	92.74	8.55	0.5939	25.27	4528.00
0.0460	92.63	8.89	0.6056	26.39	4496.00
0.0521	92.76	8.87	0.6048	26.20	4508.00

Table-6: ANL (A) + mCLP (B) in Benzene (S)

X_A	V_M (cm^2mole^{-1})	$\beta a \times 10^{-11}$ (cm^2dyne^{-1})	$L_f \times 10^{-10}$ (m)	Va (cm^3mole^{-1})	R
0.0000	92.41	7.77	0.5661	20.06	4580.00
0.0064	91.81	7.68	0.5627	21.59	4557.00
0.0128	91.67	7.73	0.5647	21.72	4547.00
0.0193	91.15	7.74	0.5649	21.69	4549.00
0.0258	90.82	7.63	0.5610	21.11	4514.00
0.0324	90.50	7.49	0.5558	20.38	4512.00

2. Graphs: The graphs of ultrasonic velocity (U) and adiabatic compressibility (βa) vs. weight fraction of amines (A) are shown in the Fig.-1 to 6.

Fig.-1: The ultrasonic velocity (U) and adiabatic compressibility (βa) vs. weight fraction of amines (A) for the system- nBA (A) + oCLP (B) in Benzene(S) at the temperature 313K

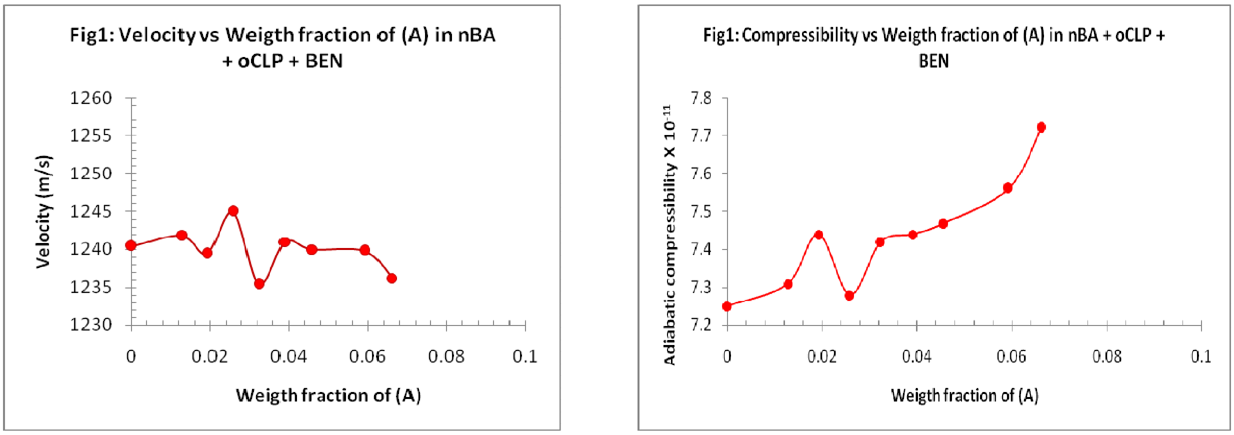


Fig.-2: The ultrasonic velocity (U) and adiabatic compressibility (βa) vs. weight fraction of amines (A) for the system- nBA (A) + pCLP (B) in Benzene (S) at the temperature 313K

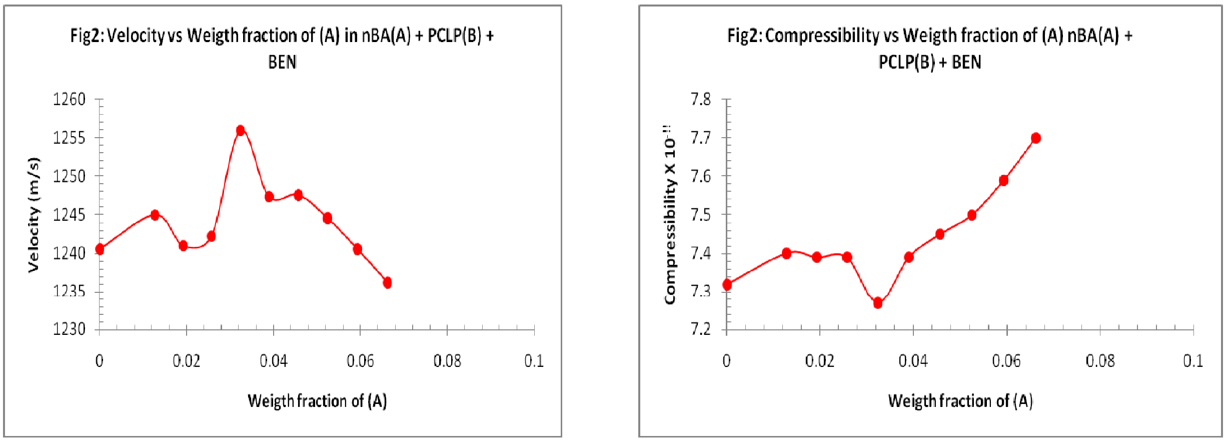


Fig.-3: The ultrasonic velocity (U) and adiabatic compressibility (βa) vs. weight fraction of amines (A) for the system- nPA (A) + oCLP (B) in Benzene (S) at the temperature 313K

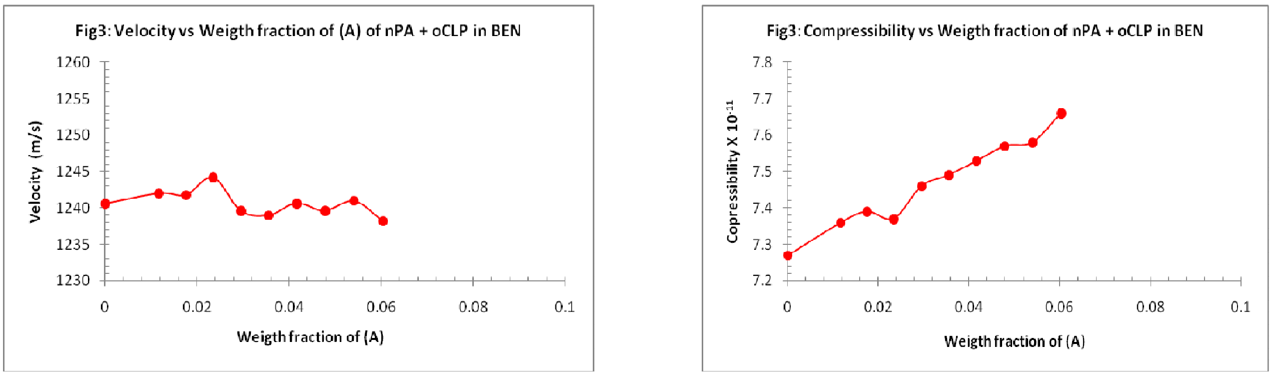


Fig.-4: The ultrasonic velocity (U) and adiabatic compressibility (βa) vs. weight fraction of amines (A) for the system- nPA (A) + pCLP (B) in Benzene (S) at the temperature 313K

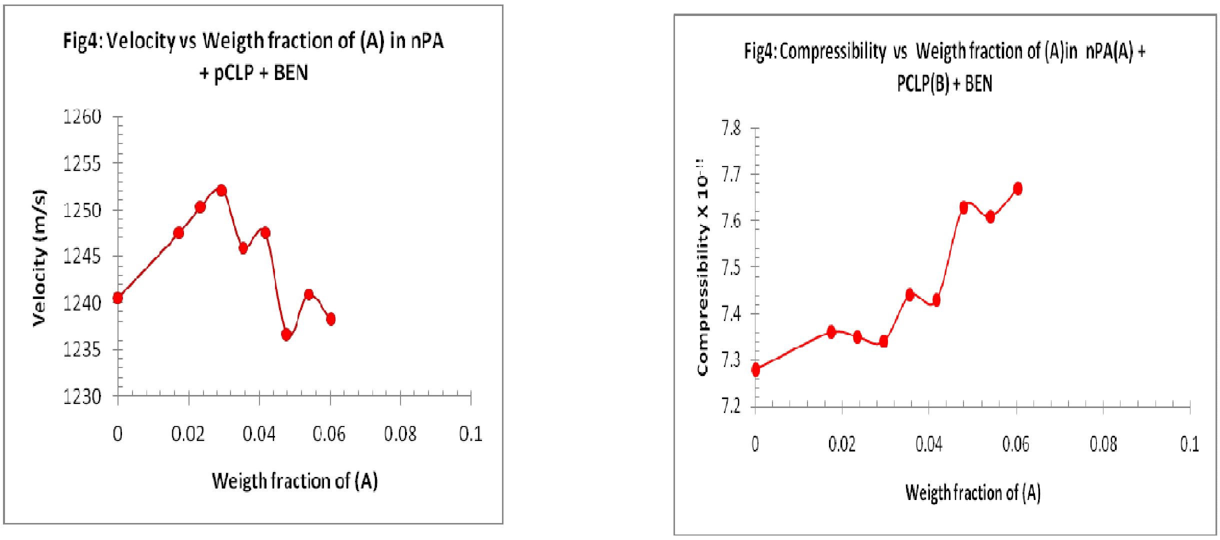


Fig.-5: The ultrasonic velocity (U) and adiabatic compressibility (βa) vs. weight fraction of amines (A) for the system- TEA (A) + mCLP (B) in Benzene (S) at the temperature 313K

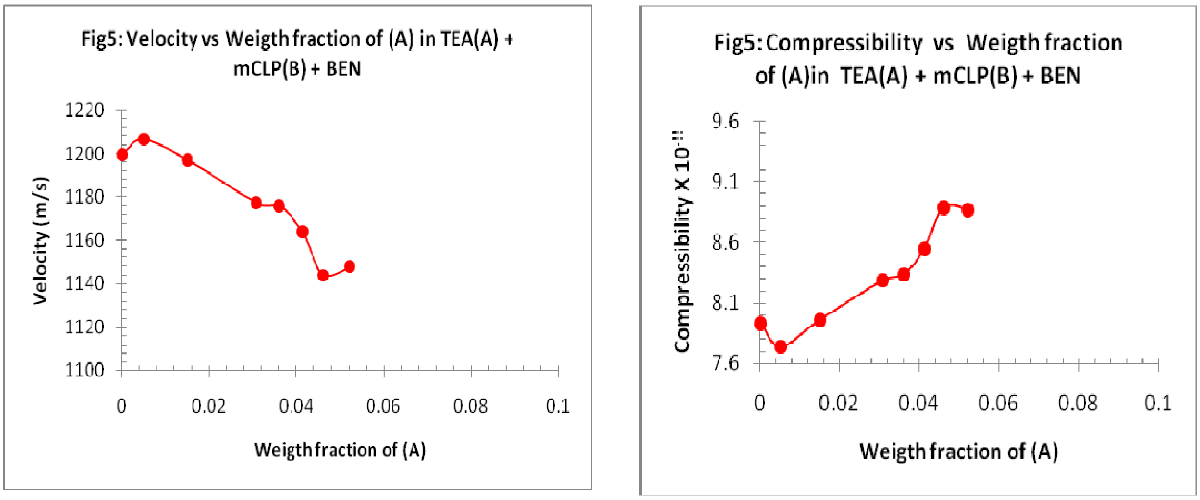
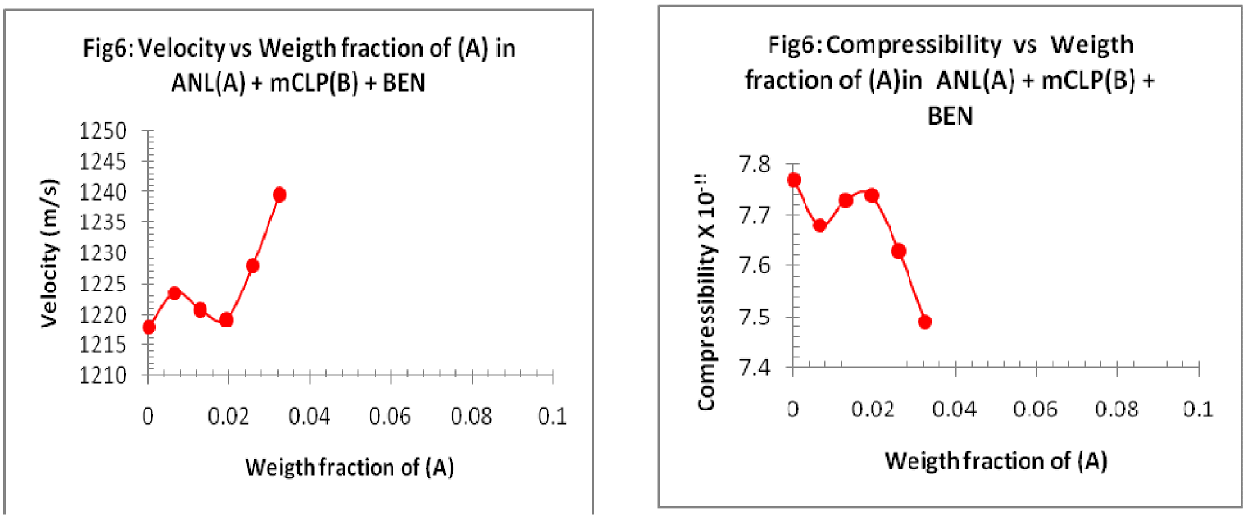


Fig.-6: The ultrasonic velocity (U) and adiabatic compressibility (βa) vs. weight fraction of amines (A) for the system- ANL (A) + mCLP (B) in Benzene (S) at the temperature 313K



CONCLUSION

The ultrasonic velocity and density have been measured experimentally for the systems presented in this paper at the temperature 313K. The variation of ultrasonic velocity and adiabatic compressibility vs. weight fraction of amines (A) shows non-linear variations and results complex formation. Molar Volume, Available Volume, Free Length and Rao's constant respectively are evaluated. Maximum, observed in ultrasonic velocity vs. concentration curve for n-Butylamine + p-chlorophenol in benzene, indicates complex formation of O-H…N type. This is supported by minimum in compressibility, Free Length and available volume at this concentration. On the other hand ultrasonic velocity curve for n-Butylamine + o-chlorophenol in benzene has slightly convex upward trend. Chlorine in the ortho-position of oCLP reduces the volume of closest approach with nBA and as such the molecular interaction is reduced between n-Butylamine + o-chlorophenol than n-Butylamine + p-chlorophenol. Similar behavior is observed in n-Propylamine + p-chlorophenol and n-Propylamine+o-chlorophenol in benzene. Trimethylamine + m-chlorophenol in benzene has convex upward trend while Aniline + m-chlorophenol has concave upward trends.

This indicates that extend of interaction of mCLP is more with Trimethylamine than Aniline. This may be due to more basicity of Trimethylamine than Aniline.

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