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## Study on association reactions for some phenolic Schiff base derived from benzoyl acetonitrile

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### ABSTRACT

The project was started by the synthesis of four Schiff bases derived from the mother compound benzoyl acetonitrile. These imines were synthesized by a standard method using ethanol solvent. Their chemical structures were confirmed previously by using chemical reagents and physical method using U.V-I.R spectra and melting point.

The main aim of the project was to study the extent association of acetonitrile benzoyl o,m and p- amino phenols by spectrophotometry using a distribution method in benzene solvent. This method gave a precise and accurate values for determination of concentrations for solute in organic and water phases using partition method.

Hence the degree of association(n) and the de association constant (K) for these Schiff bases could be estimated, discussed and interpreted.

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### KEYWORDS

Schiff base;  
Association reactions;  
Thermodynamic;  
Dipole – dipole;  
Spectrophotometry.

### INTRODUCTION

Intermolecular hydrogen bonding was know as an oldesand original method used for association of molecules. This encourage several workers to study association of molecules, by this method.

Later Hayman<sup>[2]</sup> had determined association constants of 1:1 charge transfer complexes by using spectrophotometric and partition method. This led to the believe that spectrophotometric association constant were usually smaller thanpartition method.

In 1981 Change had published several methods were used in studying association<sup>[3]</sup> of molecules. This led Yamaguchi<sup>[4]</sup> et al to determine association constants between 5-guanosine mono phosphate gel and aromatic

compounds by capillary electrophoresis. Later Kuppeas<sup>[5]</sup> had studied the intermolecular association of tetrahydrofuran-2-carboxylic acid in solution with vibrational circular dichroism. Finally Azzouz<sup>[6]</sup> and AL-Niemi had introduced for the first time the idea of ion-induced dipole as an additional method in association of substituted benzoic acids beside the intermolecular hydrogenbondings. This interesting study encourage the workers in this investigation to study association of acetonitrile benzoyl nylidene o, m and p- amino phenols, using spectrophotometric method. These molecules contain phenol groups in ortho, meta and para positions on aromatic rings.

A comparison between the degrees of association constant and (n) values ofassociation constants K are

given and discussed. This opened the ways for other subsequent works in future.

## EXPERIMENTAL

All chemicals used throughout this work were supplied from Fluka, Aldrich and Molecule companies.

They were benzoyl acetonitrile, o-amino phenol, m-amino phenol and p-amino phenol. Ethanol was bought from local market.

All Schiff bases under study were prepared by using a standard method<sup>[7]</sup>, or by mixing equimolar amounts of benzoyl acetonitrile with o,m and p-amino phenols. About 10 ml of absolute ethanol was added to any mixture, followed by refluxing the mixture for about 90 minutes, cooling and filtration of products. Recrystallization of products were performed using absolute ethanol.

Schiff bases acetonitrile benzoyl nylidene o,m and p-amino phenols were abbreviated as I, II and III respectively.

### Distribution method

This method had been applied is studying the extent of association of Schiff bases (I-III) in benzene solvent, by using similar procedure<sup>[6,7]</sup> adopted elsewhere.

Before applying the distribution method in studying the extent of association reaction in any Schiff bases using a spectrophotometric method for estimation of solute in organic phase, it was found with great importance to prepare a calibration curve for each imine solution in benzene. This required the following:-

A stock  $10^{-3}$ M solutions of imines (I-III) were prepared in benzene solvent. From this solution a range of solutions having concentrations range between  $(1-7)10^{-4}$ M were prepared. The U.V spectrum for each solution was measured at  $\lambda_{\max}$ . A plots of absorbances of Schiff bases (I-III) versus concentrations were shown in Figure 1

These showed a very good straight lines of correlation coefficient range values 0.9952-0.9986 or obeying Beer's-Lambert law. The sensitivity of the method for Schiff bases determination, as measured in terms of  $\epsilon_{\max}$  have a range of values (3306-4873) in  $\text{Liter.mole}^{-1}.\text{cm}^{-1}$ .

Hence the concentration of any Schiff bases in ben-

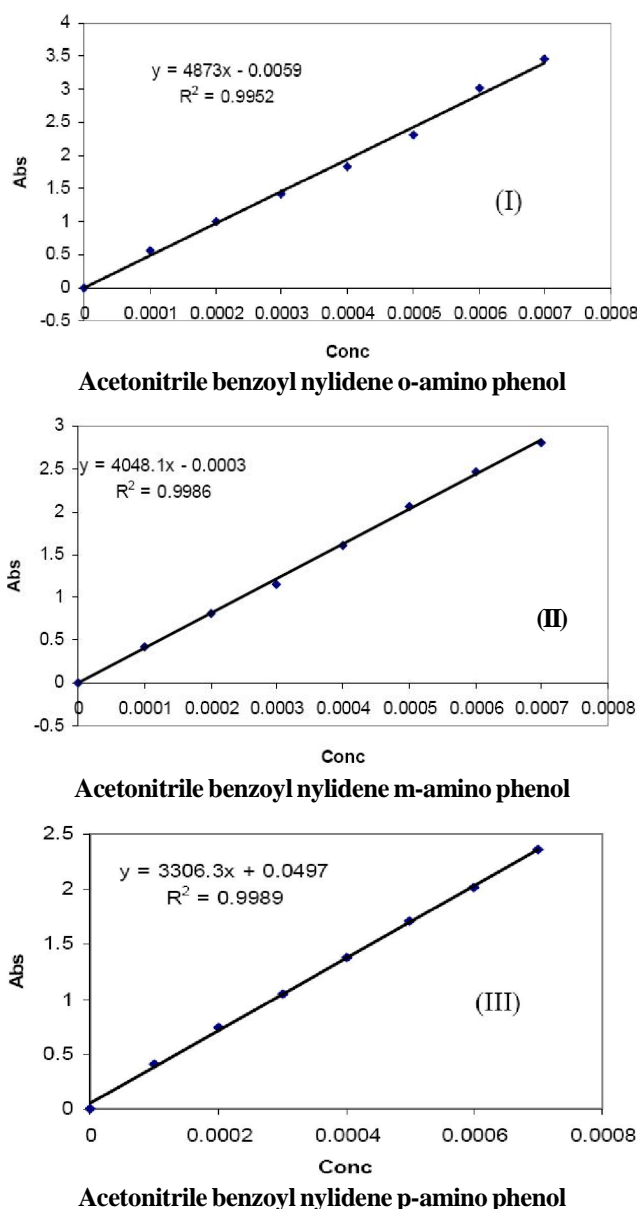


Figure 1 : Calibration curve in benzene solvent for Schiff's bases

zene ( $C_o$ ), can be estimated safely. The concentration of Schiff base in water phase ( $C_w$ ) during partition between benzene-water phases in any mixture could be calculated easily from an equation of the form:-

$$\text{Total concentration of Schiff base} = C_o + C_w$$

$C_o$  denotes to concentration of solute Schiff base in organic ( $o$ ) benzene solvent.

$C_w$  denotes to concentration of solute Schiff base in aqueous ( $w$ ) water solvent.

### Distribution method

This method had been applied in studying extent of association of Schiff bases I,II and III in benzene sol-

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vent, by using similar procedure adopted in standard or book<sup>[7]</sup> or literature<sup>[8]</sup>, as in the following:-

Each mixture was stirred for 90 minutes at tem-

ml. of 10 <sup>-3</sup> M S.B in benzene	2	4	6	8	10
ml. of benzene	18	16	14	12	10
ml. of water	20	20	20	20	20

perature range (298-333)°K. Then after each mixture was left for 15 minutes for the separation of benzene phase from aqueous phase. Finally, the concentration for any Schiff base in benzene was determined from calibration curve prepared.

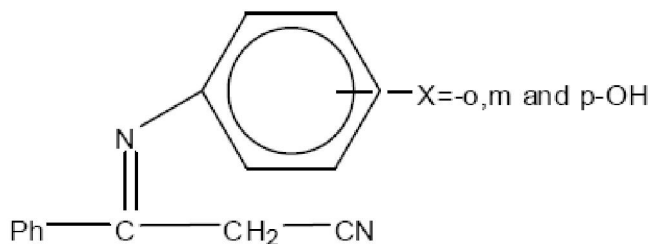
### Instrumentation

- 1 The calibration curve for U.V spectra of imines I,II and III were measured by using a computerized double beam Shimadzu 1601 using silica cell of dimensions 1x1x3cm<sup>3</sup>.
- 2 Shaker – Water bath, JEIO Tech., type BS-11, made in Korea.

## RESULTS AND DISCUSSION

The structures of Schiff bases under study, having a general formulas of the following:-

These showed that phenolic groups circulate in o,



m and p-position of amine of Schiff base molecules. These differences in structures were expected to influence on melting points and extent of association process.

Melting points of pure acetonitril benzoyl nylidene o, m and p-amino phenols Schiff bases have melting points of 67°C, 71°C, 63°C respectively. The high melting point of II, means its ability to associate or polymerize in solid state by one of association method's as hydrogen bonding or other methods.

### Distribution method

In an earlier study Wall<sup>[8]</sup> et al had applied this method for studying association of benzoic acid and its

substituents. Later Azzouz<sup>[10]</sup> et al had studied association of salicylidene 2-hydroxyaniline. These studies encourage the same last Worker<sup>[6]</sup> to deal with association of substituted benzoic acids and other acids by physical methods in benzene solvent, using the following equation:-

$$\log C_w = \frac{1}{n} \log C_o + \text{Constant} \quad (1)$$

$C_w$  = Concentration of Schiff base in water phase

$C_o$  = Concentration of Schiff base in organic phase

$n$  = Degree of association of solute

Data collected from last studies at temperature range (20-60) °C where shown in TABLES 1-3.

For each Schiff base I,II and III, the plots of  $\log C_w$  versus  $\log C_o$  at five different temperatures in the range stated, showed a very good lines of correlation coefficients ranged 0.9595-0.9951 as in Figure 1. These indicated the applicability of equation (1) on experimental association processes of Schiff bases under studying.

TABLE 4 showed association collected by distribution method for Schiff bases I,II and III in non polar benzene solvent at five temperatures. This clear the following points:-

- 1 Acetonitrile benzoyl nylidene p- amino phenol having a degrees of associations ( $n$ ) between 0.292- 1.051. In other words, Schiff base III exist as a monomer in benzene. These showed a direct relationship between absolute temperatures and the degrees of association, and agreed<sup>[6]</sup> with association of 4-carboxy benzoic acid.
- 2 Acetonitrile benzoyl nylidene m- amino phenol exist as monomer in solvent stated. These having a degrees of association range between 0.872- 1.379. A contrary to the previous finding were an inverse relationship between absolute temperatures and the degrees of association at five different temperatures listed in TABLE 4, and agreed with association<sup>[6]</sup> of 4-nitro benzoic.
- 3 Acetonitrile benzoyl nylidene o- amino phenol association results, was agreed with Schiff base acetonitrile benzoyl nylidene p- amino phenol, by showing a direct relationship between absolute temperatures and the degrees of association. A disagreement between these Schiff bases i.e III and I by observing a degrees of association range be-

TABLE 1: Distribution method of acetonitrile benzoyl nylidene o-amino phenol between water and benzene at different temperatures

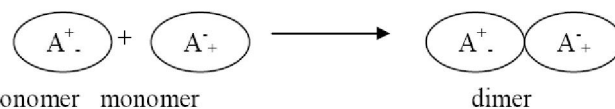
Temperature (C°)	No.	Concentration of Schiff base inwater ( $C_w \cdot 10^{-4}$ ) (mol.lit <sup>-1</sup> )	log $C_w$	Concentration of Schiff base inbenzene ( $C_o \cdot 10^{-4}$ ) (mol.lit <sup>-1</sup> )	log $C_o$	$C_o / C_w$
20	1	0.734	-4.134	1.343	-3.872	1.8297
	2	0.843	-4.074	2.084	-3.681	2.4721
	3	1.030	-3.987	2.735	-3.563	2.6553
	4	1.293	-3.888	3.491	-3.457	2.6978
	5	1.602	-3.795	4.634	-3.334	2.8908
30	1	0.997	-4.001	1.079	-3.967	1.081
	2	1.151	-3.939	1.778	-3.750	1.544
	3	1.286	-3.891	2.483	-3.605	1.929
	4	1.559	-3.807	3.228	-3.491	2.070
	5	1.808	-3.743	4.426	-3.354	2.447
40	1	1.155	-3.937	0.920	-4.036	0.797
	2	1.377	-3.861	1.552	-3.809	1.126
	3	1.537	-3.813	2.234	-3.651	1.451
	4	1.777	-3.750	2.965	-3.528	1.668
	5	2.195	-3.658	4.108	-3.386	1.869
50	1	1.305	-3.884	0.771	-4.113	0.591
	2	1.578	-3.802	1.349	-3.870	0.855
	3	1.845	-3.734	1.923	-3.716	1.042
	4	2.048	-3.689	2.742	-3.562	1.337
	5	2.560	-3.597	3.707	-3.431	1.467
60	1	1.362	-3.866	0.714	-4.146	0.524
	2	1.643	-3.784	1.285	-3.891	0.781
	3	1.898	-3.722	1.871	-3.728	0.984
	4	2.179	-3.662	2.606	-3.584	1.197
	5	2.649	-3.577	3.581	-3.446	1.352

tween 1.541- 2.477 in Schiff base I and temperatures stated. i.e Schiff base I exist as a dimer in benzene. This interesting results, encourage the workers in this investigation to search for the reason of such abnormality, or way such molecule exist as a dimer. To answer such important question, this led to consider the possible methods of association in molecule I as follower:-

- 1 By intermolecular hydrogen bonding of similar molecules. This happen due to existence of phenolic group in imineI. Hence hydrogen bonding of type O-H...O may presumably exist. This type of bonding was known in literature<sup>[1]</sup> was weakened by the elevation of temperature. Hence the degrees of association (n) must be lowered by an increase of temperatures. These finding in TABLE 4 for imine I dose not with last paragraph. Therefore, the possibility of association of I by hydrogen bonding was

no longer to exist or having a lower weight in the association process.

- 2 By dipole-dipole association method. Imine I contains a powerful electron withdrawing cyano group. This resulted to the production of two positive and negative centers in Schiff base I, or creating a dipole in it. The mechanism<sup>[9]</sup> of association by dipole-dipole method in Schiff base I as designated by A symbol, could be represented in scheme (I). The force (f) required for separation of charges in



monomer monomer

dimer

A molecule could be defined by an equation of the form:

$$F = \frac{q_1 q_2}{\epsilon r^2} \quad (1a)$$

$q_1$  = charge of positive ion  
 $q_2$  = charge of negative ion

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**TABLE 2 : Distribution method of acetonitrile benzoyl nylidenem-amino phenol between water and benzene at different temperatures**

Temperature (C°)	No.	Concentration of Schiff base inwater ( $C_w * 10^{-4}$ ) (mol.lit <sup>-1</sup> )	Log $C_w$	Concentration of Schiff base inbenzene ( $C_o * 10^{-4}$ ) (mol.lit <sup>-1</sup> )	Log $C_o$	$C_o / C_w$
20	1	1.159	-3.936	0.845	-4.073	0.729
	2	1.503	-3.823	1.337	-3.874	0.890
	3	2.065	-3.685	1.910	-3.719	0.925
	4	2.594	-3.586	2.518	-3.599	0.971
	5	2.897	-3.538	3.170	-3.499	1.094
30	1	0.883	-4.054	1.122	-3.950	1.271
	2	1.197	-3.922	1.644	-3.784	1.373
	3	1.503	-3.823	2.466	-3.608	1.641
	4	1.936	-3.713	3.170	-3.499	1.637
	5	2.228	-3.652	3.864	-3.413	1.734
40	1	0.685	-4.164	1.321	-3.879	1.928
	2	0.857	-4.067	1.981	-3.703	2.311
	3	1.191	-3.924	2.780	-3.556	2.334
	4	1.560	-3.807	3.548	-3.450	2.274
	5	1.750	-3.757	4.345	-3.362	2.483
50	1	0.404	-4.394	1.603	-3.795	3.968
	2	0.589	-4.230	2.249	-3.648	3.818
	3	0.774	-4.111	3.199	-3.495	4.133
	4	0.984	-4.007	4.130	-3.384	4.197
	5	1.183	-3.927	4.909	-3.309	4.150
60	1	0.198	-4.703	1.807	-3.743	9.126
	2	0.292	-4.534	2.547	-3.594	8.723
	3	0.451	-4.346	3.524	-3.453	7.814
	4	0.574	-4.241	4.539	-3.343	7.908
	5	0.687	-4.163	5.407	-3.267	7.870

**TABLE 3 : Distribution method of acetonitrile benzoyl nylidenep-amino phenol between water and benzene at different temperatures**

Temperature (C°)	No.	Concentration of Schiff base inwater ( $C_w * 10^{-4}$ ) (mol.lit <sup>-1</sup> )	log $C_w$	Concentration of Schiff base inbenzene ( $C_o * 10^{-4}$ ) (mol.lit <sup>-1</sup> )	log $C_o$	$C_o / C_w$
20	1	0.110	-4.959	1.824	-3.739	16.582
	2	0.485	-4.314	2.371	-3.625	4.889
	3	0.923	-4.035	2.951	-3.530	3.197
	4	1.445	-3.840	3.420	-3.466	2.367
	5	1.706	-3.768	4.074	-3.390	2.388
30	1	0.573	-4.242	1.361	-3.866	2.375
	2	0.940	-4.027	1.919	-3.717	2.041
	3	1.285	-3.891	2.588	-3.587	2.014
	4	1.774	-3.751	3.083	-3.511	1.738
	5	2.042	-3.690	3.741	-3.427	1.832
40	1	0.679	-4.168	1.256	-3.901	1.850
	2	1.079	-3.967	1.770	-3.752	1.640
	3	1.556	-3.808	2.317	-3.635	1.489
	4	1.986	-3.702	2.877	-3.541	1.449
	5	2.317	-3.635	3.467	-3.460	1.496
50	1	0.891	-4.050	1.045	-3.981	1.173
	2	1.406	-3.852	1.552	-3.809	1.104
	3	1.766	-3.753	2.109	-3.676	1.194
	4	2.265	-3.645	2.594	-3.586	1.145
	5	2.606	-3.584	3.177	-3.498	1.219

Temperature (C°)	No.	Concentration of Schiff base in water ( $C_w \cdot 10^{-4}$ ) (mol.lit <sup>-1</sup> )	log $C_w$	Concentration of Schiff base in benzene ( $C_o \cdot 10^{-4}$ ) (mol.lit <sup>-1</sup> )	log $C_o$	$C_o / C_w$
60	1	1.062	-3.974	0.873	-4.059	0.822
	2	1.462	-3.835	1.333	-3.875	0.912
	3	2.042	-3.690	1.832	-3.737	0.897
	4	2.588	-3.587	2.270	-3.644	0.877
	5	3.069	-3.513	2.716	-3.566	0.885

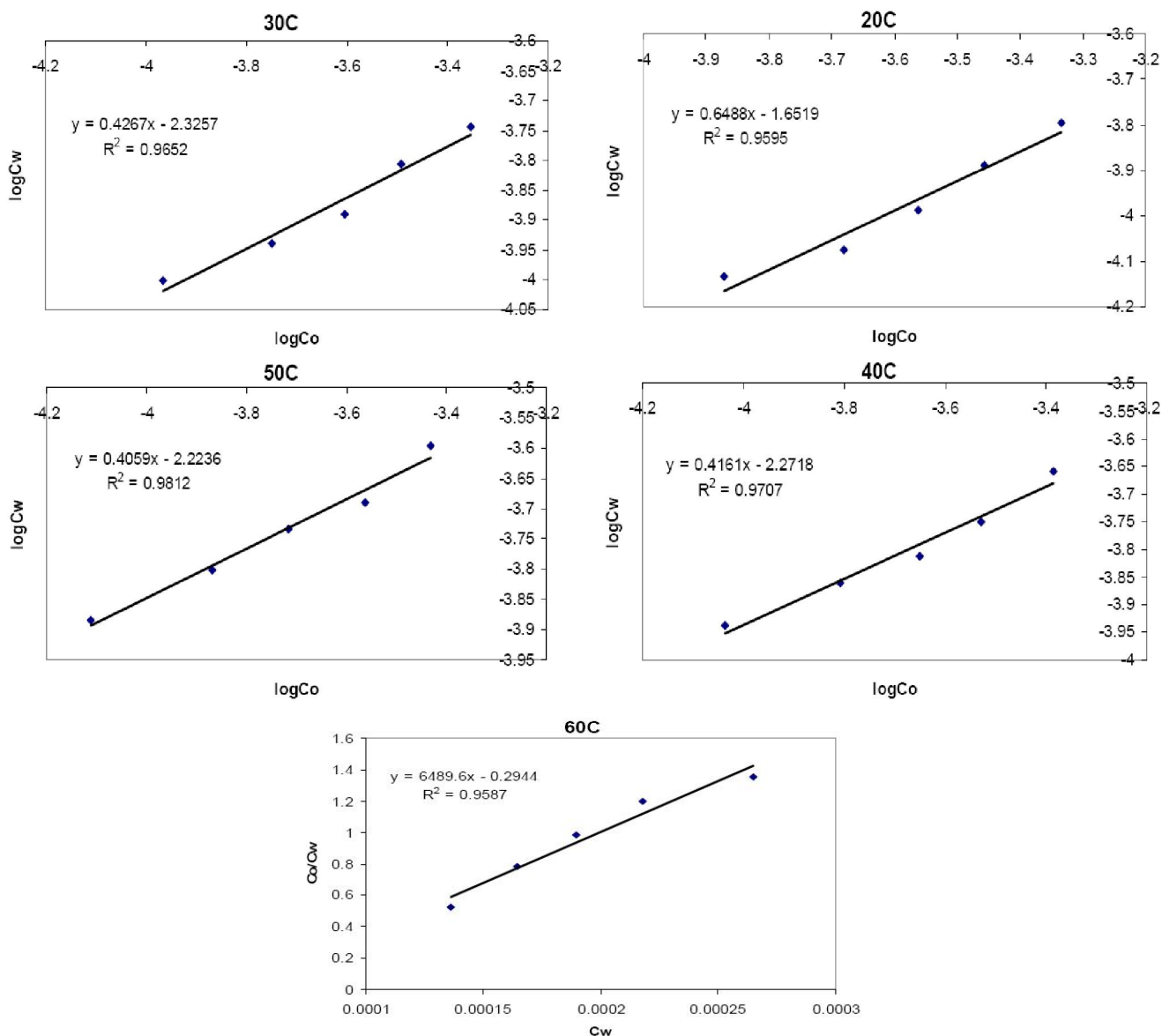


Figure 2 : Plot of  $\log C_w$  versus  $\log C_o$  for acetone nitrile benzoyl nylideneo-amino phenol

$r$  = distance between charges

$\epsilon$  = dielectric constant of the medium

By increasing the value of  $r$ , hence values of charges  $q_1$  and  $q_2$  were expected to be increased, This might resulted to an increase in the dipole moment of Schiff base monomer. The last might resulted to an increase in the degrees of association of Schiff base I by an eleva-

tion of temperatures.

Surely, the temperature as known had a great influence on many chemical reaction as stability<sup>[10]</sup> of azo dyes formation tautomerism of some benzoin<sup>[11]</sup> compounds and dyes<sup>[12]</sup> and  $pK_a$  of acidic and basic imines<sup>[13]</sup>, These studies encourage the workers to deal with such essential study.

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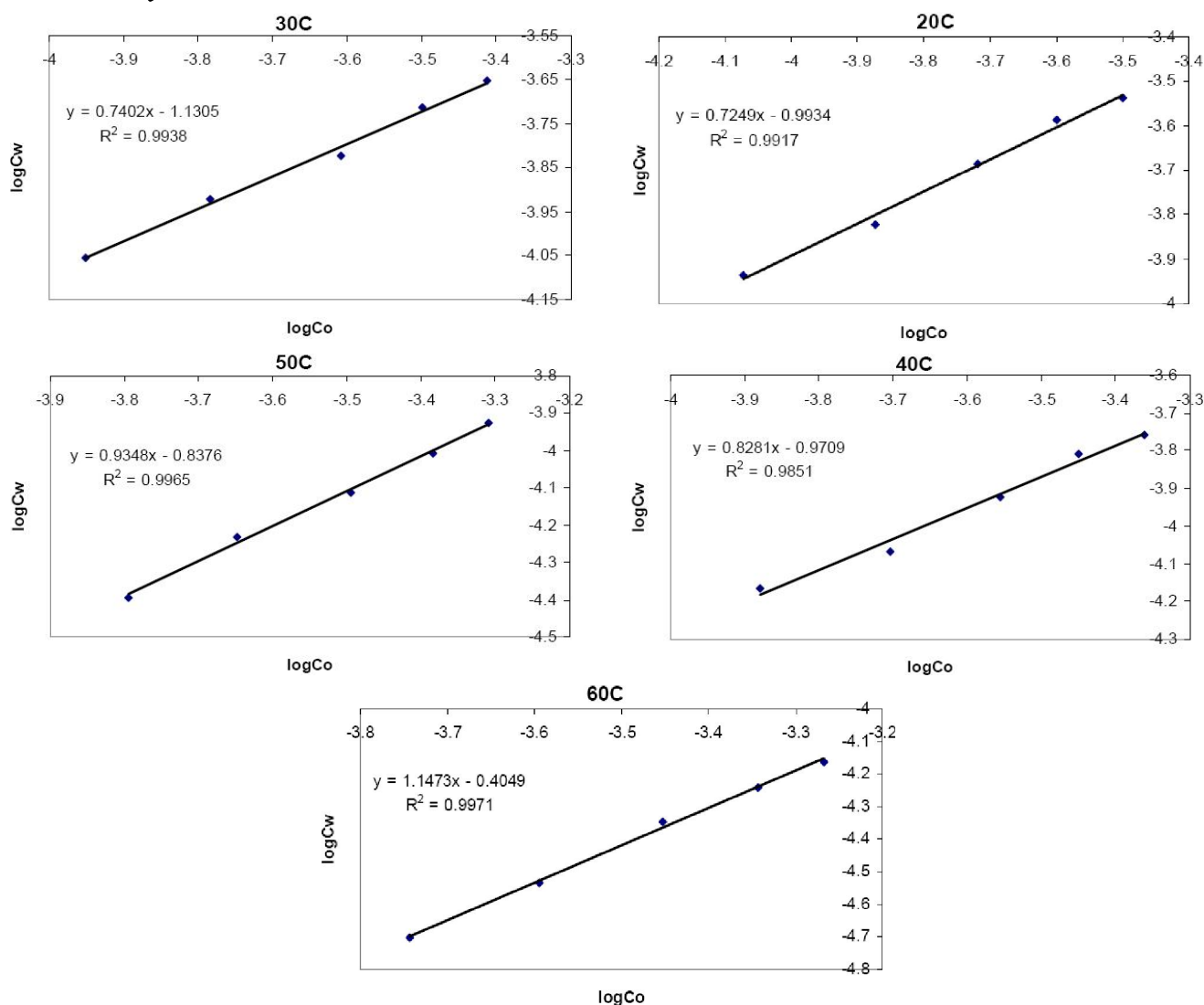
Figure 3 : Plot of  $\log C_w$  versus  $\log C_o$  for acetonitrile benzoyl nylidene m-amino phenol

TABLE 4 : Association results Schiff base I- III in benzene

Symbol	Temperature (K)	Intercept	Slope	Degree of association(n)	Correlation Coefficient
Acetonitrile benzoyl nylidene o-amino phenol	293	-1.6519	0.6488	1.541	0.9595
	303	-2.3257	0.4267	2.344	0.9652
	313	-2.2718	0.4161	2.403	0.9707
	323	-2.2236	0.4059	2.464	0.9812
	333	-2.2047	0.4037	2.477	0.9829
Acetonitrile benzoyl nylidene m-amino phenol	293	-0.9934	0.7249	1.379	0.9917
	303	-1.1305	0.7402	1.351	0.9938
	313	-0.9709	0.8281	1.206	0.9851
	323	-0.8376	0.9348	1.070	0.9965
	333	-0.4049	1.1473	0.872	0.9971
Acetonitrile benzoyl nylidene p-amino phenol	293	7.9676	3.4228	0.292	0.9361
	303	0.7031	1.2766	0.783	0.9916
	313	0.6408	1.2294	0.963	0.9935
	323	-0.2004	0.9640	1.037	0.9932
	333	-0.1281	0.9511	1.051	0.9941

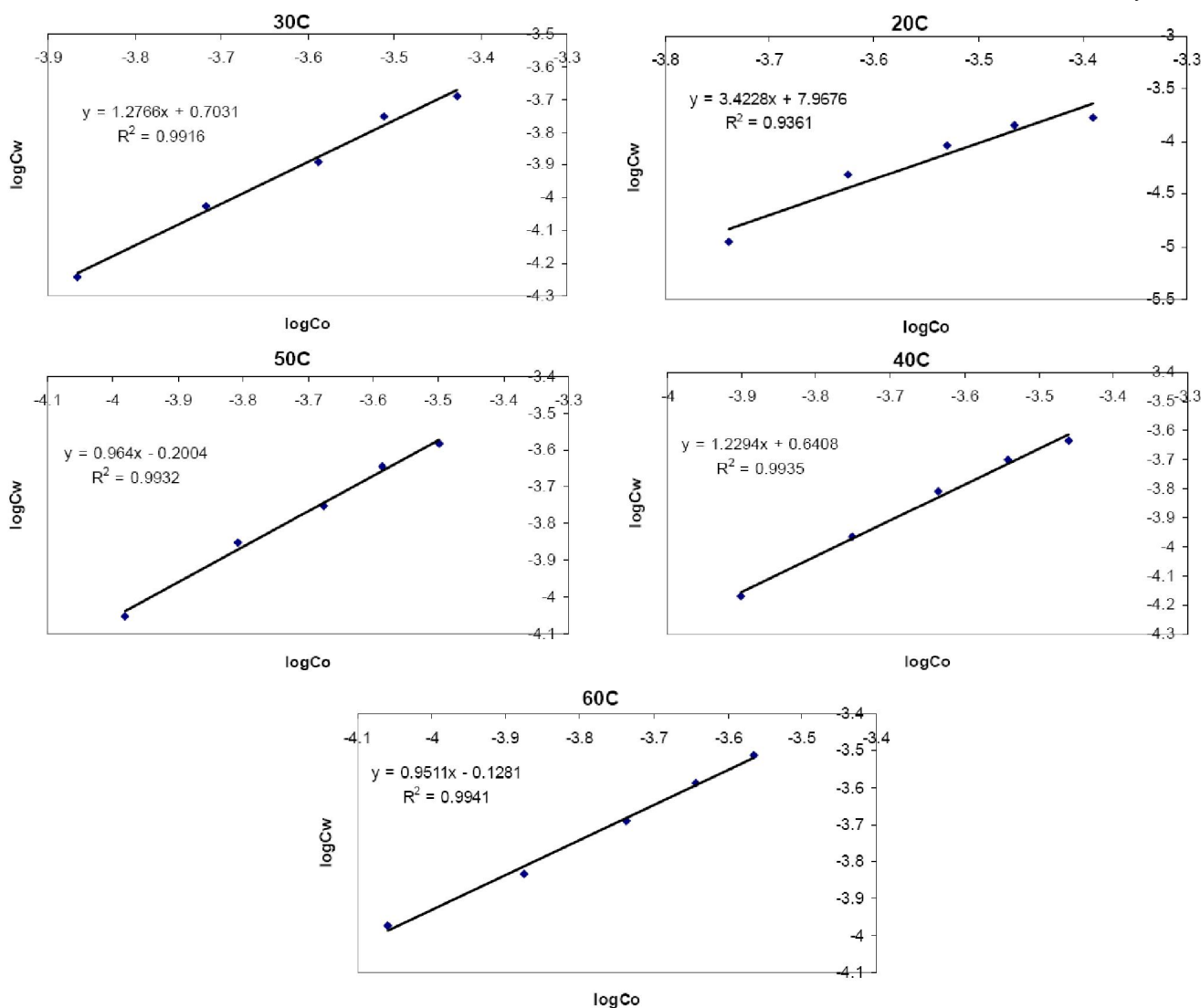


Figure 4 : Plot of  $\log C_w$  versus  $\log C_o$  for acetonitrile benzoyl nylidene p-amino phenol

The thermodynamic of association represent the various energies forms associated with conversion of with conversion of dimer Schiff base I to the corresponding monomer i.e dimer  $\xrightleftharpoons{K_2} 2$ monomer. These required the evaluation of thermodynamic parameters, namely the  $\Delta G^\circ$ ,  $\Delta H^\circ$  and  $\Delta S^\circ$ . The last required of the evaluation of equilibrium constants for the reaction outlined above. Hence the following equation<sup>[6]</sup> was used for the dimeric form of reactant.

$$\frac{C_o}{C_w} = K_1 + \frac{2K_1^2 C_w}{K_2} \quad (2)$$

$K_1$  = distribution coefficient (constant)

$K_2$  = equilibrium constant.

Form equation (2) the plot of  $C_o/C_w$  versus  $C_w$  at temperature rang stated showed a straight lines of cor-

relation coefficient values of 0.9829.

Hence  $K_1$  and  $K_2$  were evaluated. Now when  $\ln K_2$  values for the de association reaction of Schiff base I were plotted versus the values of  $T^{-1}$  form Table. This showed a straight line of negative slope of  $R^2$  value equal (0.991), as in figure.

The thermodynamic parameters for the de association reaction of Schiff base I were evaluated from standard equations (3-5) of forms:-

$$\Delta G^\circ = -RT \ln K_2 \quad (3)$$

$$\ln K_2 = \text{Constant} - \frac{\Delta H^\circ}{RT} \quad (4)$$

$$\Delta G^\circ = \Delta H^\circ - T\Delta S^\circ \quad (5)$$

TABLE 5 showed the thermodynamic parameters calculated form  $K_2$  equilibrium constant i.e Schiff base



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I dimer  $\leftrightarrow$  2 monomer. It was cleared from TABLE 5 that all positive signs of  $\Delta G^\circ$  calculated for Schiff base I dimer to their corresponding monomer was difficult and non spontaneous.

This process was highly expected, due to dipole-dipole forces among any molecules of dimer, which results to an increase in the stability of the latter if compared with its monomers.

The enthalpy of reaction  $\Delta H^\circ$  was calculated from Van Hoff equation shown in equation (4). An Excel computer programme was also used in the evaluation

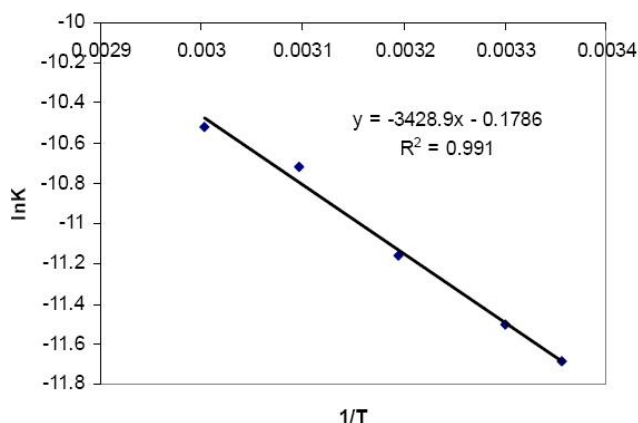


Figure 5 : The relationship between  $\ln K_2$  versus  $T^{-1}$  for the de association reaction of Schiff base (I)

TABLE 5 : Equilibrium constant values for the de association reaction of Schiff base I at various temperatures and thermodynamic

Symbol	Temperature (K)	$\ln K_2$	$\Delta H^\circ$ J.mol <sup>-1</sup>	$\Delta H^\circ$ J.mol <sup>-1</sup>	$\Delta G^\circ$ J.mol <sup>-1</sup>	$\Delta G^\circ$ J.mol <sup>-1</sup>	$\Delta S^\circ$ J.mol <sup>-1</sup> .K <sup>-1</sup>	$\Delta S$ J.mol <sup>-1</sup> .K <sup>-1</sup>
(I)	298	-11.686	28510.4		28952.9		-1.48	
	303	-11.503	28527.8		28977.7		-1.48	
	313	-11.158	28571.5	28508.1	29036.3	28974.4	-1.48	-1.48
	323	-10.716	28297.4		28776.9		-1.48	
	333	-10.521	28633.6		29128.0		-1.48	

### CONCLUSION

- 1 There Schiff bases prepared from reactions of benzoyl acetonitril with o,m- and p-amino phenols by standard method. They are abbreviated as I,II and III respectively.
- 2 The structures of Schiff base I, II and III were confirmed by physical method, namely by using UV and IR spectra and melting points.
- 3 A spectrophotometric method was applied for the determination of concentration for Schiff bases I,II

of  $\Delta H^\circ$  for Schiff base I at five different temperatures. TABLE 5 illustrates the following fact:

The near values of  $\Delta H^\circ$  calculated for Schiff base I at different five temperatures means the non dependence of  $\Delta H^\circ$  at narrow temperature rang between 298 and 333K.

$\Delta S^\circ$  values for the reaction stated were calculated from equations.

Theoretically, the de dimerization reaction of Schiff base I was directed toward the 2 monomers i.e  $\Delta S^\circ$  values must have a positive signs.

$$\Delta S^\circ = S_2 - S_1$$

$S_2$  = entropy of monomer product

$S_1$  = entropy of dimer Schiff base I

All  $\Delta S^\circ$  values at temperatures listed in TABLE 5 had a negative signs of single value of 1.48 J.mol<sup>-1</sup>.K<sup>-1</sup>. These results were in contrary to theoretical values stated before. This only happen if values  $S_1 > S_2$ . This only happen if monomers produced from the de dimerization reaction recombined by dipole – dipole association method outlined before. This agreed with increasing order of product monomers and lowering  $S_2$  values as compared with  $S_1$ , leading to a negative values of  $\Delta S^\circ$  parameter.

and III in benzene solvent.

- 4 The degrees of association of I, II and III were evaluated by distribution method at temperature rang (293-333)K in benzene – water medium. These showed a dimeric monomeric and monomeric existence of I, II and III respectively in non polar benzene solvent.
- 5 Schiff base I was able to dimerize in benzene by acid of dipole – dipole association method.
- 6 The association constant (k) for the dimerization reaction of I were evaluated at temperatures range stated.

7 The thermodynamic parameters, namely  $\Delta G^\circ$ ,  $\Delta H^\circ$ ,  $\Delta S^\circ$  for the de dimerization reaction of Schiff base I were interpreted and discussed.

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