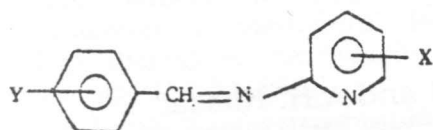


structure skeleton for these compounds is given as follows:



Where:

X	Y	Ligand
H	o-OH	(L ₁)
p-CH ₃	o-OH	(L ₂)
o-CH ₃	o-OH	(L ₃)
H	o,p-OH	(L ₄)
H	m,p-OH	(L ₅)

The electronic absorption spectra were recorded using a Perkin-Elmer Lambda (4B) spectrophotometer. The solvents used were of spectroquality; benzene, chloroform, acetone, ethanol, methanol, dimethylformamide, acetonitrile, dimethylsulphoxide and water. The solvents were selected to show a wide variation in dielectric constant (ϵ), refractive index (n) and hydrogen bonding capacity. The data were treated using the Gauss-Jordan elimination method.

3. Results and discussion

The IR spectral bands of the prepared Schiff's base compounds are given in Table 2.

The bands in the range 1280-1285 cm^{-1} were ascribed to the phenolic $\nu_{\text{C-O}}$ stretching vibration [20]. The absence of ν_{OH} bands in the range 3650-3590 cm^{-1} was attributed to the existence of intramolecular hydrogen bonding [21]. The strong band which appeared at about 1570-1540 cm^{-1} was assigned to the $\nu_{\text{C=N}}$ stretching mode [4].

The electronic absorption spectra of the above-mentioned compounds, in selective organic solvents, are given in Table 3. Eight absorption bands: A, B, C, D, E, F, G, and H appeared in the ranges 209-240 nm, 250-270 nm, 270-298 nm, 303-339 nm, 340-358 nm, 360-378 nm, 380-401 nm, and 407-470 nm, respectively. The bands A, B, and C were assigned to $\pi \rightarrow \pi^*$ transitions of aromatic rings [22], while the band D was attributed to $n \rightarrow \pi^*$ transitions of the pyridine ring [23] and the intramolecular charge transfer [6]. The band E was assigned to $n \rightarrow \pi^*$ transition of CH=N group [7]. The electronic absorption bands F and G are considered as the charge transfer interaction through the whole molecule [13, 24]. However the band H was attributed to the hydrogen-bonding interaction between the solvent and the dissolved solute [25, 26].

Table 1 Melting point, color and elemental analysis of benzylidene 2-(azomethine pyridine) Schiff's base compounds.

Ligands	m.p. °C	Color	Elemental analysis (Calculated)		
			C	H	N
o-hydroxybenzylidene-2-(azomethine pyridine) L ₁	65	Yellow	72.5 (72.7)	5.3 (5.1)	14.3 (14.1)
o-hydroxybenzylidene-2-(azomethine-4-methylpyridine), L ₂	90	Yellow	73.8 (73.6)	5.5 (5.7)	13.5 (13.2)
o-hydroxybenzylidene-2-(azomethine-6-methylpyridine), L ₃	80	Orange	73.6 (73.6)	5.5 (5.7)	13.1 (13.2)
2,4-dihydroxybenzylidene-2-(azomethine pyridine) L ₄	Charring	Orange	67.4 (67.3)	4.5 (4.7)	13.0 (13.1)
3,4-dihydroxybenzylidene-2-(azomethine pyridine) L ₅	Charring	Orange	67.5 (67.3)	4.7 (4.7)	12.9 (13.1)

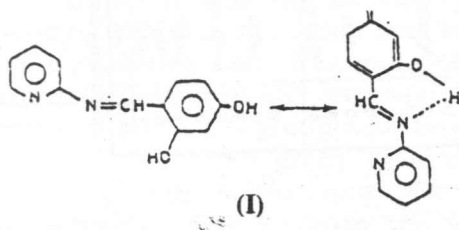
Table 2. Assignment of some characteristic infrared bands of some benzylidene-2-(azomethine pyridine) Schiff's base compounds

Ligand	ν_{OH}	$\nu_{\text{C=Npy}}$	$\nu_{\text{C=Nazomethine}}$	$\nu_{\text{C-O}}$	$\delta_{\text{C-OH}}$
L ₁	3440	1610	1550	1280	1185
L ₂	3430	1600	1540	1280	1195
L ₃	3450	1612	1550	1282	1185
L ₄	3450	1612	1550	1282	1140
L ₅	3450	1615	1530	1280	1160

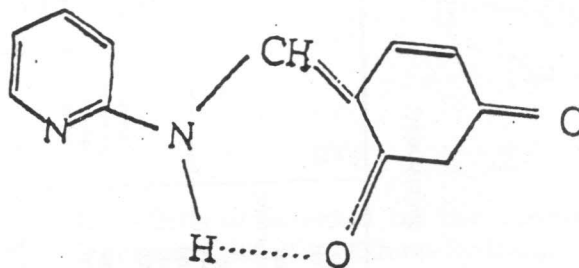
ν_{OH} : infrared-stretching frequency for phenolic OH group.
 $\nu_{\text{C=Npy}}$: infrared stretching frequency for C=N group in pyridine ring.
 $\nu_{\text{C=Nazomethine}}$: infrared stretching frequency for azomethine group.
 $\nu_{\text{C-O}}$: infrared-stretching frequency of phenolic C-O.
 $\delta_{\text{C-OH}}$: infrared-bending frequency for phenolic OH group.

By comparing the electronic absorption spectra of *o*-hydroxybenzylidene-2-azomethine pyridine (parent) with other investigated compounds, the following observations were made:

1. The electronic absorption spectra of dihydroxybenzylidene compounds showed an additional broad band in the range (495-580 nm), assigned quinonoid tautomer (I) [18-26].
2. The spectral bands E and H of 2, 4 - dihydroxybenzylidene compound show red and blue shifts, respectively, relative to the corresponding bands of *o*-hydroxybenzylidene (parent) compound. The red shift is due to the formation of intramolecular hydrogen bonding between the *o*-OH of benzyl ring and the lone pair of electrons of the nitrogen atom [10-23] which consequently decreased the ability of the formation of intermolecular hydrogen bonding. The quinonoid tautomer (I) of 2, 4-dihydroxy-compound [18-26] renders the behavior of the compound to be a proton acceptor toward the solvent, where a blue shift takes place. The appearance of a band around 290 nm for 2,4-dihydroxy-compound and its absence in the parent compound, is due to the overlapping of π -electrons of CH=N-with the π - electrons of benzylidene [18-27]part since, they lie in the same plane as a result of the formation of intramolecular hydrogen bonding. In view of this argument, the tautomeric structure I for 2,4-dihydroxy-compound can be represented as follows:



3. The appearance of a new band with high intensity due to $n \rightarrow \pi^*$ transitions of C=O group [18], and the absence of bands within the range 340-358 nm and 407-470 nm in water solvent, for 2,4-dihydroxy-compound, may be attributed to the formation of diketo-amine compound II [27].



(II)

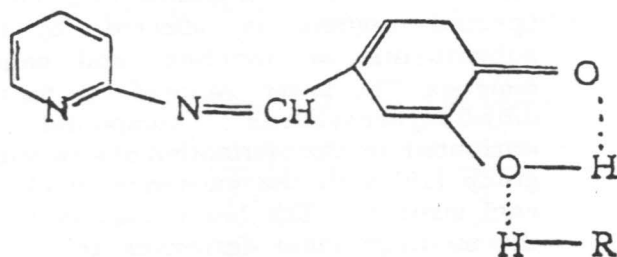
4. The hydroxyl group in the meta and para positions relative to the CH=N group at benzylidene part of the 3,4-dihydroxy-compound prevents the formation of intramolecular hydrogen bonding. The weak spectral band E of the 3,4-dihydroxy-compound shows a slight shift, and a blue shift relative to parent compound and the 2,4-dihydroxy derivative, respectively. This may be attributed to the formation of intermolecular hydrogen bonding between the solvent and the lone pair of electrons on the nitrogen atom of CH=N- group [7]. The electronic absorption band H of the 3,4-dihydroxy-compound exhibits a blue shift relative to the parent compound. This shift arises from the formation of the intramolecular hydrogen bonding between the carbonyl group and the adjacent hydroxyl group in the benzylidene part [18, 27]. So this compound behaves as a proton acceptor towards the solvent. This argument leads to the following structure:

Table (3) Effect of Solvents on the Electronic Absorption Spectra of some benzylidene-2-(azomethine pyridine) Schiff bases Compounds

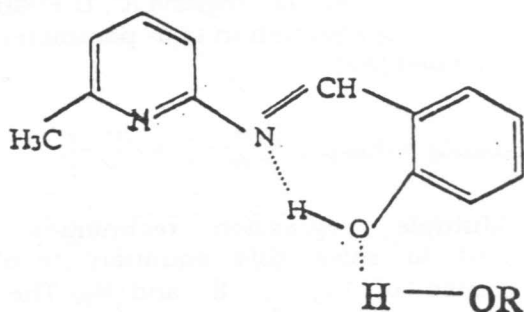
Compisolvents	Benzene	CHCl ₃	Acetone	EtOH	MeOH	DMF	CH ₃ CN	DMSO	H ₂ O	Bands
L ₁	-	245(v.w)	241(w),231(w),214(s)	209(s)	210(s)	225,211(v.vw)	234(s),266(w)	-	231(s)	A
	270(s)	265(w)	270.4(w)	269	267.4(m)	268.4(s)	267.6(m)	269(m)	257(s)	B
	-	298.3(m)	-	-	-	-	-	-	288(s)	C
	306(s)	313.1(sh)	331.8,320.9	301.7(s)	301.7(s)	303.6(s)	303.2(s)	308.8(s)	-	D
	350.7(s)	340.9(m)	345.6(s)	348.3(s)	348.4(m)	348.3(s)	342.5(m)	348.5(m)	341.8(sh.w)	E
	-	354(m)	368.5(s)	366.4(m)	371.9(w)	364.2(m)	364.5(m)	371(w)	-	F
	391.6(sh)	392(sh)	395(sh)	387.4(sh)	383.6(w.sh)	381.2(sh)	394.5(m)	393.4(sh)	382(sh)	G
	468(sh)	461(sh)	459.7(sh)	453.3(sh)	451(sh)	465.8(sh)	451.7(sh)	458.1(sh)	-	H
-	-	578.6	-	-	-	-	-	-	-	-
L ₂	-	240.2(s)	241,225,212(w)	240.2(w)	213(s)	-	227(s)	250.6	218.9(s)	A
	286.7(v.vw)	261.8(w)	265(w)	261.8(w)	270(m)	-	277.8(w)	270.8(sh)	-	B
	295.6(v.vw)	290.8(m)	286(sh)	292.8(w)	-	-	293.4(w)	296.6(m)	286.8	C
	336.4(v.w)	336.9(m)	319(s)	337.9(m)	301.8(sh)	335.4(w),323.8(s)	338.6(s)	306.6(s)	-	D
	346.4(sh.w)	321(v.w)	347(s)	357.7(w)	344.2(s)	356.8(w),343.3(w)	354.8(w)	349.5(s)	-	E
	366.8	353.5(v.w)	-	366.2(m)	363.9(s)	366.4(s)	364.5(w)	346(m)	-	F
	401(s)	366.4(m)	393.8(m)	391.6(m)	-	391.6(m)	383.8(sh)	-	352.6(b)	G
	461(sh.w)	391.5(sh)	451.7(sh)	461(sh)	448.4(sh)	461(sh)	458.1(sh.w)	458.5(sh)	-	H
-	464.9(sh)	579(w.b)	-	-	-	-	669.8(b)	-	-	
L ₃	-	242.0(s)	213.4(s)	229.2(s)	219(s)	-	223.6(s)	269.4(s)	228.2(s)	A
	-	267.6)	-	-	267.8(s)	-	267(s)	285	-	B
	293.4(w)	288.6(w)	-	293.4,280.4	-	-	-	323.2(s)	290.6(s)	C
	-	320(s)	320.4(w)	307.2(s)	307(s)	322.2(s)	307.6(s)	348(s)	-	D
	345.1(v.v.vw)	343.2(m)	347.2(s)	345(s)	345.2(s)	348.6(s)	345.2(s)	371(s)	-	E
	363.4(s)	361.2	-	370.9,360.4	370.9(s)	367.7(s)	361.3(s)	389.4(w)	-	F
	393.6(m)	-	391(m)	389(w)	389(w)	390.6(w)	-	458.5(sh)	384(b)	G
	471.1(sh)	467.8(sh)	458(w.sh)	445.2(sh)	440.4(sh)	459.7(sh)	467.8	-	-	H
-	571.2(b.w)	-	-	-	-	-	-	-	-	
L ₄	212(w)	-	-	-	-	-	-	-	227	A
	265(m)	-	-	-	-	-	-	254.8	-	B
	293.4(s)	288.2(s)	-	292.8(s)	290(s)	-	291(s)	296.6(s)	287.6(s)	C
	-	328.5(wb)	333.4(w)	331.2	-	322.5(w)	332.2(w)	-	317.9(sh)	D
	345.0(v.w)	343.4(v.vw)	346.4(s)	353(s)	350.7(s)	348.3(s)	345.1(s)	351.5(v.w)	-	E
	368.8(v.w)	365.6(s)	-	361.2(v.w)	-	-	-	-	-	F
	385.8(sh)	-	371.9(s)	-	377.5(w)	372(s)	-	370.9(s)	380.8(sh)	G
	422.6(sh)	-	420.8(s)	-	-	423.2(s)	413(s)	407.8(s)	-	H
-	-	561.5(w)	506.6(sh)	-	-	534(w)	551.8(b)	495.9(s)	-	
L ₅	-	-	212.4(w),211.3(s)	235(s)	210(s),230(w)	-	239.2(w),216.2(s)	-	219(m)	A
	-	-	267.4	-	-	-	-	251(s)	-	B
	-	-	-	280.4(w)	280.6(w)	-	-	285.2(s)	275.6(w)	C
	-	-	331(s),319.4(w),309.5(sh)	342.9(w)	342.9(w)	321(s)	343(w)	-	-	D
	-	-	343.8(s)	-	-	336.4(v.w)	-	-	-	E
	-	-	-	-	-	342.9(w)	-	343.4(s)	343(w)	F
	-	-	386.8(m)	-	-	-	380.6(w.sh)	386.3(s)	383(s)	G
	-	-	439(s)	438.8(sh)	425.8(sh)	449.2(sh)	448.4(sh)	451.7(sh)	422.6(sh)	H
-	-	577.7(sh)	-	-	577.4(sh.w)	582.6	564.7(w)	580.6(v.vw)	-	

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Abbreviation S: strong; m: medium; sh: shoulder; vw: very weak and b: broad



(III)



(Solvent)

(IV)

5. The position of the methyl group in the pyridine ring affects the electronic absorption bands of compounds L_2 and L_3 . The *p*-methyl pyridine L_2 - compound exhibits red and blue shifts in regions D, E and H, respectively, relative to the corresponding bands of the *o*-hydroxybenzylidene compound. This arises from the electronic donor power of the methyl group to increase the charge density on CH=N-group and $n \rightarrow \pi'$ transitions of the pyridine ring. This leads to increasing the energy for the "n" orbital of the CH=N-group with a consequent decrease in the energy for the $n \rightarrow \pi'$ transition. Accordingly, the tendency for the formation of intramolecular hydrogen bonding between the *o*-OH group in the benzyl ring and the nitrogen atom of the CH=N-group increases. However, the electronic absorption spectra of *o*-methyl pyridine in regions E and H exhibit a blue shift (but the H band shows a red shift in benzene, chloroform and acetone) relative to the parent and *p*-methyl pyridine compounds. The red shift is attributed to the formation of intramolecular hydrogen bonding [10]. The blue shift may be due to the existence of intramolecular and intermolecular [10,28] hydrogen bonding between the *o*-OH group and the nitrogen atom of the CH=N group, and the *o*-OH group of the solute and the solvent, respectively. This leads to the following structure:
6. The effect of solvents on the electronic absorption bands of the *o*-hydroxybenzylidene, 3,4-dihydroxybenzylidene and *o*-methylpyridine compounds consists of slight displacements, and does not involve a fundamental change of the spectra. The band (E) exhibits a red shift from a non-polar to a polar solvent for 2,4-dihydroxybenzylidene and *p*-methyl-pyridine-compounds. This may be attributed to that the excited state is more polar than the ground state [29,30]. So polar solvents stabilized the excited state: by lowering the energy of the excited state, the band appears at a longer wavelength. Some exceptions are observed due to the formation of intermolecular hydrogen bonding. [31]
7. In presence of water as a solvent, the investigated compounds absorbed at shorter wavelengths than that in alcohol (methanol and ethanol) due to the formation of external hydrogen bonding. The blue shifted in bands are going from in nonpolar to polar solvent [28,31].
- The effect of the main physical properties of the solvents, namely, the dielectric constant, ϵ , and the refractive index, n , on the electronic spectral bands was studied. Table 4 shows the solvents utilized in each case. The spectra of each sample in various solvents were studied and the peak locations were visually determined from spectral charts, as in Table 3. The coefficients K_1 , K_2 and the

intercept (ν_{vapor}) were calculated for each peak in different spectral regions (C, D E, and H). The following equation in two-parameter form was applied [32]

$$\nu_{\text{(solution)}} = \nu_{\text{(vapor)}} - K_1 \frac{2\varepsilon - 2}{2\varepsilon + 1} - K_2 \frac{2n^2 - 2}{2n^2 + 1} \quad (1)$$

Multiple regression techniques were applied to solve this equation to obtain estimates of ν_{vapor} , K_1 and K_2 . The data analyses are given in terms of three variables ε , n and ν_{solution} . For a linear equation of the form shown in Eq. (1), the least-squares criterions require that the square of the sum of the errors can be minimized. In this case, the residual error, S , is.

$$S = \sum_{i=1}^m \left[\nu_{\text{solution}} - \nu_{\text{vapor}} + K_1 E_i + K_2 N_i \right]^2, \quad (2)$$

where

$$E = \frac{2\varepsilon - 2}{2\varepsilon + 1}, \quad N = \frac{2n^2 - 2}{2n^2 + 1}.$$

Differentiating S with respect to ν_{vapor} , K_1 and K_2 and setting each partial derivative equal to zero, three normal equations for ν_{vapor} , K_1 and K_2 were obtained. The correlation coefficient (r) between the two parameters, x , y was calculated as follows:

$$r = \frac{n \sum xy - \sum x \sum y}{\sqrt{(n \sum x^2 - (\sum x)^2) (n \sum y^2 - (\sum y)^2)}}, \quad (3)$$

where x represents ε or n and y corresponds to ν_{solution} .

The magnitude of r determines the strength of the relationship between pairs of variables. The sign of r tells whether y tends to increase or decrease with x . The value of r must satisfy the inequality $-1 \leq r \leq 1$, i.e. $r = \pm 1$ if the points of the scatter line lie on a straight line. The calculated ν_{vapor} , K_1 , K_2 , $r(\nu, n)$, $r(\nu, \varepsilon)$ and S for the organic compounds are collected in Table 5.

4. Conclusions

1. The ν_{vapor} of the compounds, in different spectral regions is affected by the substituents at pyridine and benzyl moieties. The lower value of ν_{vapor} for the dihydroxybenzylidene compound is attributed to the formation of a carbonyl group [18] with the existence of a keto-enol structure. The lower ν_{vapor} value for the methylpyridine derivatives relative to that of the parent compound is attributed to the electron donating power of methyl group in different positions of the pyridine ring [10]. The ν_{vapor} values of the *p*-methylpyridine compound exhibits a blue shift (based on band H) and red a shift (based on band E) relative to the *o*-methylpyridine compound. This is due to the ability of the formation of intramolecular hydrogen bond (26,28). This is more pronounced in the case of the *p*-methylpyridine compound. However the intermolecular hydrogen bond is more in the case of the *o*-methylpyridine compound.
2. Both the dielectric constant and the refractive index of the solvents are of considerable importance in explaining the effect of solvents on the electronic spectral data. The refractive index of the solvent is of major importance in controlling the behavior of the 2,4-dihydroxybenzylidene, the 3,4 dihydroxybenzylidene, the *o*-hydroxybenzylidene, the *p*-methylpyridine and the *o*-methylpyridine compounds based on spectral regions (H,C), E,C and D, respectively. Based on E and C spectral regions for the 2,4-dihydroxybenzylidene and the *o*-methylpyridine compounds, respectively, the dielectric constant is the major factor. However, in the case of the *o*-hydroxybenzylidene and the *o*-methylpyridine compounds, based on spectral region H, both the refractive index and dielectric constant are of a considerable effect. The low correlation coefficient value (r) for some different spectral regions of investigated compounds, Table 5, indicate that the spectral peak shift did not arise

from the effect of the dielectric constant and refractive index of the used solvent. This shift can be explained by solute solvent hydrogen bonding [10, 28] as

previously discussed in examples I, II, III and IV.

Table 4. ϵ , n , E and N for solvents.

Solvent	ϵ	H	E	N
Benzene	2.3	1.501	0.464	0.455
CHCl ₃	4.7	1.443	0.712	0.419
Acetone	20.7	1.359	0.929	0.361
Et OH	24.3	1.361	0.940	0.362
Me OH	32.6	1.329	0.955	0.338
DMF	36.7	1.427	0.960	0.409
CH ₃ CN	37.5	1.344	0.961	0.350
DMSO	48.9	1.478	0.970	0.441
H ₂ O	78.5	1.333	0.981	0.341

Where: ϵ is the dielectric constant, n is the refractive index,
 $E = (2\epsilon - 2)/(2\epsilon + 1)$, $N = (2n^2 - 2)/(2n^2 - 1)$.

Table 5. K_1 , K_2 , ν_{vapor} , S , and r for some benzylidene-2-(azomethine pyridine) Schiff's base compounds.

Compounds	ν_{vapor}	K_1	K_2	$r(\nu, \epsilon)$	$R(\nu, n)$	$S \times 10^6$	$S^2 \times 10^6$	Bands
	38612.7	156.0	-3537.9	0.21	0.28	2.90	0.57	B
	-	-	-	-	-	-	-	C
L ₁	33293.7	144.3	-2215.5	0.13	0.17	3.30	0.64	D
	30794.7	-1.5	-4867.3	-0.35	-0.90	0.06	0.09	E
	23236.5	367.8	-4449.9	0.64	0.80	0.21	0.16	H
	-	-	-	-	-	-	-	B
L ₂	39096.2	-593.5	-10998.0	-0.42	0.73	0.97	0.37	C
	-	-	-	-	-	-	-	D
	30012.8	-921.8	-1881.0	0.27	-0.05	1.10	0.37	E
	22461.8	112.7	-2046.4	0.32	0.42	0.35	0.21	H
	38705.5	-552.2	-2239.0	0.25	0.66	0.02	0.06	B
	36939.5	-1262.3	-3640.4	0.70	-0.47x10 ⁻²	0.03	0.07	C
L ₃	38112.2	178.0	-16248.8	-0.25	0.82	1.2	0.41	D
	30546.9	-780.1	-2468.3	0.45	0.14	0.07	0.10	E
	29747.1	119.0	-2573.3	0.43	0.59	0.22	0.16	E
	21969.2	1277.4	-3125.0	0.60	0.54	1.19	0.39	H
	-	-	-	-	-	-	-	B
L ₄	38212.8	-1235.8	-7365.0	-0.05	0.53	0.42	0.24	C
	28334.1	1627.5	1819.2	0.24	0.03	1.56	0.47	D
	30231.0	1124.6	-1333.6	0.56	-0.23	0.41	0.23	E
	13090.5	2042.7	2149.2	-0.14	0.50	8.88	1.22	H
	-	-	-	-	-	-	-	B
L ₅	28835.8	10159.8	-8738.7	0.25	0.84	0.09	0.02	C
	-	567.6	231.8	0.14	0.13	0.04	0.08	D
	28477.1	-1373.2	1793.0	0.23	0.85	0.42x10	0.32x10 ²	E
	18496.6	9279.1	-12375.9	0.13	0.74	0.82	0.34	H

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