

Synthesis, spectral correlations and biological activities of some (*E*)-[4-(substituted benzylidene amino)phenyl] (phenyl) methanones

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ABSTRACT

A series of Schiff's bases have been synthesized by fly-ash:*p*-toluenesulfonic acid (fly-ash:PTS) catalyzed microwave assisted oxidative coupling of arylamines and substituted benzaldehydes under solvent-free condition. The synthesized Schiff's bases are characterized by their physical constants, analytical and spectroscopic data. The group frequencies of imines such as IR (ν , cm^{-1}) and NMR (δ , ppm) spectral data was correlated with Hammett substituent constants, F and R parameters using single and multi-linear regression analysis. The antimicrobial activities of these Schiff's bases have been studied using Bauer-Kirby method.

Keywords: Schiff's bases; Fly-ash:PTS; IR spectra; NMR spectra; Hammett correlation

1. INTRODUCTION

The formation of carbon-nitrogen double bond plays important role in organic synthesis. This can be achieved by the reaction of aldehyde and amines in acidic medium which leads to synthesis of imines (or) Schiff's bases [1]. Imines are compound containing the functional group ($-\text{CH}=\text{N}-$) and they are generally called azomethine. Many Schiff's bases are known to be medicinally important and are used to design medicinal compounds [2]. They are well known intermediated for the preparation of azetidinone [3], thiazolidinone [4], formazone [5], arylacetamide [6], metal complexes [7-9] and many other derivatives [10-11]. Many reagent have been used for the synthesis of optically active imines such as Lewis acids [12], MnO_2 [13], CaO [14], ZnCl_2 [15], $\text{P}_2\text{O}_5:\text{SiO}_2$ [16], infrared [17], ultrasound radiation [18] and fly-ash: H_2SO_4 [19] with microwave irradiation. Schiff bases possess various biological activities such as anti-inflammatory [20], antibacterial [21], antifungal [22], anti-tubercular [23], anticancer [24], antimalarial [25] and anti-tumour [26], etc. Schiff bases are important intermediate for the synthesis of some bioactive compound such as β -lactams [27-29].

The spectroscopic data are useful for prediction of ground state molecular equilibration such as *s-cis* and *s-trans* conformers of chalcones, *cis* and *trans* isomers of alkenes, alkynes,

imines, ω -bromo acyl esters [30-46]. The reactivity of spectral functional group frequencies were predicted with Hammett correlation. Suresh *et al.* [19] have studied the substituent effects of some 4-methoxyphenylimines by spectral correlations. Thirunarayanan and his coworkers [47-49] have investigated the spectral study and antimicrobial activities of some trifluoromethylimines.

The effect of substituents on some symmetrical diimines has been studied by Thirunarayanan [50]. The spectral correlation and antimicrobial activity of some benzylidene pyridine-2-amine and benzylidene-3-nitrobenzenamine have been investigated by Sakthinathan *et al.* [51,52]. Recently, Mayavel *et al.* [43] have studied the solvent-free synthesis, spectral correlation and antimicrobial activities of some (*E*)-*N*-(substituted benzylidene)-9-ethyl-9*H*-carbazole-3-amines. Within the above view, there is no report available for synthesis, spectral correlations and antimicrobial activities of (*E*)-[4-(substituted benzylideneamino)-phenyl](phenyl)methanone in the past.

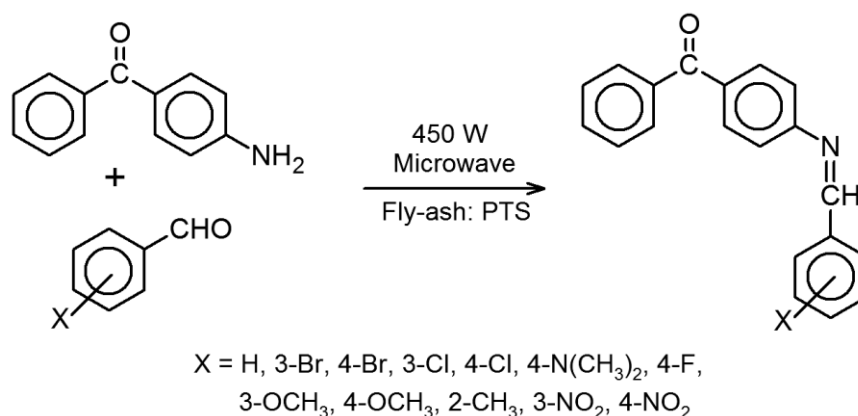
Therefore the authors have taken efforts to synthesis some (*E*)-[4-(substituted benzylideneamino) phenyl] (phenyl) methanones by fly-ash:PTS catalyzed condensation of 4-aminobenzophenone with substituted benzaldehydes to study the substituent effect by correlation analysis using Hammett substituent constants. The biological activities of these imines derivatives have been studied using Bauer-Kirby [53] method.

2. EXPERIMENTAL

2. 1. Materials and methods

All chemicals used in this work were purchased from Sigma-Aldrich Chemical Companies. Melting points of all imines have been determined in open glass capillaries on SUNTEX melting point apparatus and are uncorrected. Infrared spectra (KBr, 4000-400 cm^{-1}) have been recorded on Avatar-300 Fourier transform spectrophotometer (Thermo Nicolet, USA). The NMR spectra of all imines were recorded in Bruker AV 400 NMR spectrometer operating at 400 MHz frequency for recording ^1H NMR spectra and 100 MHz for ^{13}C NMR spectra in CDCl_3 solvent using TMS as internal standard.

2. 2. Synthesis of imines



Scheme 1. Synthesis of (*E*)-[4-(substituted benzylideneamino)phenyl](phenyl)methanones.

An appropriate equimolar quantities of aryl amines (2 mmol), substituted benzaldehyde (2 mmol) and fly-ash:PTS (0.5 g) have been taken in borosil tube and tightly capped. This mixture was subjected to microwave irradiation for 6–8 minutes in a microwave oven at 450 W [Samsung, GW73BD model, 230 V, 50 Hz, 100 N/750 W (ICE-705)] (Scheme 1). The progress of the reaction was monitored by TLC. After completion of the reaction, added 10 mL dichloromethane, the organic layer has been separated which on evaporation yields the solid product. The solid was purified by recrystallized using methanol. The analytical, physical constants and spectral data of all imines are presented in Table 1.

Table 1. Analytical, physical constants and spectral data of (E)-[4-(substituted benzylidene amino)phenyl](phenyl)methanones.

Entry	X	MF	MW	Yield (%)	m.p. (°C)	IR (cm ⁻¹)		¹ H NMR (δ, ppm)	¹³ C NMR (δ, ppm)		Mass (m/z)
						ν _{C=N}	ν _{C=O}	δ _{CH}	δ _{C=N}	δ _{C=O}	
1	H	C ₂₀ H ₁₅ NO	285	80	109–111	1589.46	1623.63	8.466	161.95	195.51	285 (M ⁺) 286 (M ¹⁺)
2	3-Br	C ₂₀ H ₁₄ BrNO	363	84	158–160	1591.92	1641.36	8.108	166.63	194.79	363 (M ⁺)
3	4-Br	C ₂₀ H ₁₄ BrNO	363	82	130–132	1592.12	1620.64	8.419	160.39	195.30	363 (M ⁺)
4	3-Cl	C ₂₀ H ₁₄ ClNO	319	78	106–108	1577.77	1635.64	8.435	160.10	195.99	319 (M ⁺) 320 (M ¹⁺)
5	4-Cl	C ₂₀ H ₁₄ ClNO	319	80	88–90	1581.63	1631.78	8.436	160.35	190.99	319 (M ⁺) 320 (M ¹⁺)
6	4-N(CH ₃) ₂	C ₂₂ H ₂₀ N ₂ O	328	85	138–140	1573.91	1637.78	8.335	161.42	195.36	328 (M ⁺)
7	4-F	C ₂₀ H ₁₄ FNO	303	79	82–84	1575.84	1627.92	8.423	160.30	195.43	303 (M ⁺)
8	3-OCH ₃	C ₂₁ H ₁₇ NO ₂	315	81	90–92	1586.60	1621.98	8.467	160.01	192.30	316 (M ¹⁺)
9	4-OCH ₃	C ₂₁ H ₁₇ NO ₂	315	82	96–98	1583.56	1635.64	8.399	161.04	195.37	316 (M ¹⁺)
10	2-CH ₃	C ₂₁ H ₁₇ NO	299	80	104–106	1587.42	1633.71	8.755	160.55	195.48	299 (M ⁺)
11	3-NO ₂	C ₂₀ H ₁₄ N ₂ O ₃	330	78	108–110	1585.49	1639.49	8.572	158.82	195.88	330 (M ⁺)
12	4-NO ₂	C ₂₀ H ₁₄ N ₂ O ₃	330	80	140–142	1589.34	1637.56	8.583	159.01	190.31	330 (M ⁺)

3. RESULTS AND DISCUSSION

3. 1. Spectral linearity

In the present study the spectral linearity of synthesized imines has been studied by evaluating the substituent effect on ν_{C=N} and ν_{C=O} (cm⁻¹). The proton chemical shift (δ, ppm) of CH=N and carbon chemical shift of C=N and C=O have been correlated with Hammett substituent constants F and R parameters using single and multi-linear regression analysis.

3. 1. 1. IR spectral study

The recorded infrared ν_{C=N}, ν_{C=O} stretching frequencies (cm⁻¹) of the synthesized imines have been presented in Table 1. These data are correlated [19,30-52] with Hammett substituent constant F and R and Swain-Lupton's [54] parameters.

In this regression analysis the structure parameter correlation Hammett equation have been employed as shown in eqn. (1).

$$\nu = \rho\sigma + \nu_0 \quad \dots (1)$$

where ν_0 is the frequency for the parent member series.

The assigned $\nu_{C=N}$ and $\nu_{C=O}$ stretching frequency (cm^{-1}) of the imine derivatives were correlated with various Hammett substituent constants F and R parameters are presented in Table 2. The correlation of $\nu_{C=N}$ (cm^{-1}) frequency of imines with Hammett substituent constant F and R parameter is found to be fail with positive ρ values except F. The resonance conjugated the structure of the imine is shown in Fig. 1.

Table 2. Results of statistical analysis of IR (ν , cm^{-1}) of C=N, C=O, NMR (δ , ppm) of CH, C=N and C=O of (*E*)-[4-(substituted benzylideneamino)phenyl](phenyl)methanones with Hammett substituent constants, F and R parameters.

Frequency	Const.	r	I	ρ	s	n	Correlated derivatives
$\nu_{C=N}$	σ	0.753	1583.59	6.741	5.75	12	H, 3-Br, 4-Br, 3-Cl, 4-Cl, 4-N(CH ₃) ₂ , 4-F, 3-OCH ₃ , 4-OCH ₃ , 2-CH ₃ , 3-NO ₂ , 4-NO ₂
	σ^+	0.872	1584.37	5.013	5.29		
	σ_I	0.759	1584.04	1.525	6.44		
	σ_R	0.811	1587.82	16.24	5.10		
	F	0.792	1585.47	-2.35	6.42		
	R	0.826	1587.93	11.67	5.03		
$\nu_{C=O}$	σ	0.860	1631.84	2.744	7.322	12	H, 3-Br, 4-Br, 3-Cl, 4-Cl, 4-N(CH ₃) ₂ , 4-F, 3-OCH ₃ , 4-OCH ₃ , 2-CH ₃ , 3-NO ₂ , 4-NO ₂
	σ^+	0.824	1632.39	-1.251	7.361		
	σ_I	0.862	1629.48	7.791	7.158		
	σ_R	0.795	1633.43	5.978	7.275		
	F	0.708	1627.94	6.113	7.256		
	R	0.720	1632.86	2.160	7.381		
δ_{CH}	σ	0.901	8.443	0.046	0.160	10	H, 4-Br, 3-Cl, 4-Cl, 4-N(CH ₃) ₂ , 4-F, 3-OCH ₃ , 4-OCH ₃ , 3-NO ₂ , 4-NO ₂
	σ^+	0.901	8.448	0.037	0.159		
	σ_I	0.839	8.481	-0.090	0.159	12	H, 3-Br, 4-Br, 3-Cl, 4-Cl, 4-N(CH ₃) ₂ , 4-F, 3-OCH ₃ , 4-OCH ₃ , 2-CH ₃ , 3-NO ₂ , 4-NO ₂
	σ_R	0.903	8.497	0.238	0.150	10	H, 4-Br, 3-Cl, 4-Cl, 4-N(CH ₃) ₂ , 4-F, 3-OCH ₃ , 4-OCH ₃ , 3-NO ₂ , 4-NO ₂
	F	0.901	8.483	-0.090	0.159		
	R	0.903	8.500	0.178	0.149		
	σ	0.902	161.01	-0.928	2.072	11	H, 4-Br, 3-Cl, 4-Cl, 4-N(CH ₃) ₂ , 4-F, 3-OCH ₃ , 4-OCH ₃ , 2-CH ₃ ,

$\delta_{C=N}$							3-NO ₂ , 4-NO ₂
	σ^+	0.901	160.90	-0.470	2.083	10	4-Br, 3-Cl, 4-Cl, 4-N(CH ₃) ₂ , 4-F, 3-OCH ₃ , 4-OCH ₃ , 2-CH ₃ , 3-NO ₂ , 4-NO ₂
	σ_I	0.902	161.57	-1.793	2.063	11	H, 4-Br, 3-Cl, 4-Cl, 4-N(CH ₃) ₂ , 4-F, 3-OCH ₃ , 4-OCH ₃ , 2-CH ₃ , 3-NO ₂ , 4-NO ₂
	σ_R	0.900	160.60	-1.369	2.085	10	4-Br, 3-Cl, 4-Cl, 4-N(CH ₃) ₂ , 4-F, 3-OCH ₃ , 4-OCH ₃ , 2-CH ₃ , 3-NO ₂ , 4-NO ₂
	F	0.903	161.64	-2.033	2.048	11	H, 4-Br, 3-Cl, 4-Cl, 4-N(CH ₃) ₂ , 4-F, 3-OCH ₃ , 4-OCH ₃ , 2-CH ₃ , 3-NO ₂ , 4-NO ₂
	R	0.904	160.60	-0.953	2.085	10	4-Br, 3-Cl, 4-Cl, 4-N(CH ₃) ₂ , 4-F, 3-OCH ₃ , 4-OCH ₃ , 2-CH ₃ , 3-NO ₂ , 4-NO ₂
$\delta_{C=N}$	σ	0.857	194.64	-1.720	1.950	12	H, 3-Br, 4-Br, 3-Cl, 4-Cl, 4-N(CH ₃) ₂ , 4-F, 3-OCH ₃ , 4-OCH ₃ , 2-CH ₃ , 3-NO ₂ , 4-NO ₂
	σ^+	0.829	194.43	-0.937	1.975		
	σ_I	0.789	195.24	-2.418	2.002		
	σ_R	0.795	194.05	-1.687	2.051		
	F	0.823	195.13	-1.968	2.032		
	R	0.796	194.05	-1.189	2.051		

r = correlation coefficient, I = intercept, ρ = slope, s = standard deviation, n = number of correlated derivatives.

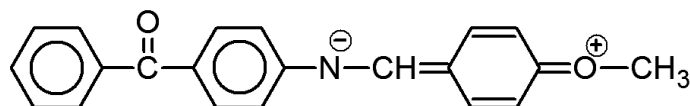


Fig. 1. The resonance-conjugative structure of imine.

The assigned the $\nu_{C=O}$ stretches were correlated with various Hammett substituent constants F and R parameter are presented in Table 2. The correlation of $\nu_{C=O}$ (cm^{-1}) frequency of imines with Hammett substituent constant F and R parameters is found to be fail with positive ρ values except σ^+ . Single parameter correlation of $\nu_{C=N}$ and $\nu_{C=O}$ frequencies of imines failed with Hammett substituent constants while seeking the multi-linear correlation, these frequencies correlated satisfactory with inductive, resonance and Swain-Lupton constants [54] and correlated equations are given in (2-5).

$$\nu_{C=N} (\text{cm}^{-1}) = 1590.14(\pm 3.704) - 5.347(\pm 0.756)\sigma_I + 18.365(\pm 7.370)\sigma_R \quad \dots (2)$$

(R = 0.906, n = 12, P > 95)

$$\nu_{C=N} (\text{cm}^{-1}) = 1590.51(\pm 3.375) - 6.109(\pm 0.687)F + 12.646(\pm 4.739)R \quad \dots (3)$$

(R = 0.906, n = 12, P > 95)

$$\nu_{\text{C=O}} (\text{cm}^{-1}) = 1626.34(\pm 4.420) - 5.021(\pm 3.881)\sigma_{\text{I}} + 17.280(\pm 9.424)\sigma_{\text{R}} \quad \dots (4)$$

(R = 0.904, n = 12, P > 95)

$$\nu_{\text{C=O}} (\text{cm}^{-1}) = 1630.44(\pm 5.092) + 5.742(\pm 0.987)F + 1.250(\pm 0.150)R \quad \dots (5)$$

(R = 0.902, n = 12, P > 95)

3. 1. 2. ¹H NMR spectral study

The ¹H NMR spectral of the imines derivatives under the investigation have been recorded in deuteriochloroform solution employing tetramethylsilane (TMS) as internal standard. The signals of the imine protons have been assigned and are presented in Table 1. In nuclear magnetic resonance spectra, the ¹H or the ¹³C chemical shift (δ , ppm) depend on the electronic environment of the nuclei concerned. The chemical shifts have been correlated with reactivity parameters. Thus the Hammett equation has been used in the form as shown in eqn. (6).

$$\delta = \delta_0 + \rho\sigma \quad \dots (6)$$

where δ_0 is the chemical shift of the corresponding parent compound.

The assigned proton chemical shift (ppm) of imines have been correlated with various Hammett constants, F and R parameters. The result of statistical analysis are presented in Table 2. The proton chemical shift (δ ppm) with Hammett σ , σ^+ , σ_{R} , F and R constants is satisfactorily. The remaining Hammett constant σ_{I} were found to be fail with positive ρ values except σ_{I} and F excluding 3-Br, 2-CH₃. This shows that the normal substituent effect operates in all system. The failure in correlation is attributed to the conjugated structure shown in Fig. 1. The authors think that, it is worthwhile to seek multiple correlations involving either σ_{I} and σ_{R} constant (or) Swain-Luptons F and R parameters. The correlation equation for CH proton chemical shift (δ , ppm) are given in eqns. (7 and 8).

$$\delta_{\text{CH}} (\text{ppm}) = 8.588(\pm 0.106) - 0.211(\pm 0.116)\sigma_{\text{I}} + 0.322(\pm 0.112)\sigma_{\text{R}} \quad \dots (7)$$

(R = 0.946, n = 12, P > 95)

$$\delta_{\text{CH}} (\text{ppm}) = 8.564(\pm 0.101) - 0.150(\pm 0.015)F + 0.201(\pm 0.012)R \quad \dots (8)$$

(R = 0.944, n = 12, P > 95)

3. 1. 3. ¹³C NMR spectra

Physical organic chemist and researches have been made extensive study of ¹³C NMR spectra for a large number of ketones, styrenes, styryl ketones and keto-epoxides. They have studied linear correlations of the chemical shifts (ppm) of C _{α} , C _{β} and C=O carbons with Hammett σ constants in alkenes, alkynes, acid chlorides and styrenes. In the present study, the chemical shifts (ppm) of imine C=N and C=O carbons have been assigned and are presented in Table 1. Attempts have been made to correlate these chemical shift (δ , ppm) with Hammett substituent constants, field and resonance parameters, with the help of single and multiple-regression analysis to study the reactivity through the effect of substituent.

The chemical shift (δ , ppm) observed for the $\delta_{\text{C=N}}$ have been correlated with Hammett constants and the results of statistical analysis are presented in Table 2.

The $\delta_{\text{C=N}}$ chemical shift (δ , ppm) gave satisfactory correlation with Hammett σ constant F and R parameters along with negative ρ values excluding (σ , σ_{I} , F, 3-Br, σ^+ , σ_{R} , R, H, 3-Br)

substituent. The chemical shift observed by $\delta_{C=O}$ carbon gave poor correlation with Hammett σ constants F and R parameters with negative ρ values.

This is due to the reason stated earlier and it is associated with the resonance conjugated structure as shown in Fig. 1. The author, think that it is worthwhile to seek multiple correlation involving all σ_I , σ_R , F and R parameters. This is given in the following correlation eqns. (9-12).

$$\delta_{C=N} \text{ (ppm)} = 161.26(\pm 1.536) - 1.504(\pm 0.969) \sigma_I - 0.772(\pm 0.057) \sigma_R \quad \dots (9)$$

(R = 0.990, n = 12, P > 95)

$$\delta_{C=N} \text{ (ppm)} = 161.38(\pm 1.431) - 1.837(\pm 0.750)F - 0.662(\pm 0.019)R \quad \dots (10)$$

(R = 0.992, n = 12, P > 95)

$$\delta_{C=O} \text{ (ppm)} = 194.96(\pm 1.48) - 2.09(\pm 0.87) \sigma_I - 0.85(\pm 0.012) \sigma_R \quad \dots (11)$$

(R = 0.930, n = 12, P > 95)

$$\delta_{C=O} \text{ (ppm)} = 194.76(\pm 1.41) - 1.69(\pm 0.71)F - 0.92(\pm 0.013)R \quad \dots (12)$$

(R = 0.928, n = 12, P > 95)

3. 2. Antimicrobial activities

3. 2. 1. Antibacterial sensitivity assay

Table 3. Antibacterial activity of (E)-[4-(substituted benzylideneamino)phenyl](phenyl)methanones.

Entry	X	Zone of inhibition (mm)				
		Gram-positive			Gram-negative	
		<i>B. subtilis</i>	<i>M. luteus</i>	<i>S. aureus</i>	<i>E. coli</i>	<i>P. aeruginosa</i>
1	H	–	–	–	–	2
2	3-Br	–	–	–	2	–
3	4-Br	–	–	–	–	3
4	3-Cl	–	–	1	–	2
5	4-Cl	1	1	–	–	1
6	4-N(CH ₃) ₂	–	–	1	–	–
7	4-F	1	–	1	2	–
8	3-OCH ₃	–	–	1	–	–
9	4-OCH ₃	–	–	–	2	–
10	2-CH ₃	–	–	2	–	–
11	3-NO ₂	–	–	–	1	–
12	4-NO ₂	–	–	–	–	1
Amacilin		8	3	6	4	1
DMSO		–	–	–	–	–

Antibacterial sensitivity assay has been performed by using disc diffusion [53] technique. In each petri plate about 0.5 mL of the test bacterial sample has been spread uniformly over the solidified Mueller Hinton agar using sterile glass spreader. Then the disc with 5 mm diameter made up of Whatman No. 1 filter paper, impregnated with the solution of the compound have been placed on the medium using sterile forceps. The plates have been incubated for 24 h at 37 °C by keeping the plates upside down to prevent the collection of water droplets over the medium. After 24 h, the plates have been visually examined and the diameter values of the zone of inhibition were measured. Triplicate results have been recorded by repeating the some procedure.

The antibacterial screening effect of synthesized Schiff's bases is shown in Fig. 3. The antibacterial activities of all the synthesized imines have been studied against three Gram-positive pathogenic strains *Bacillus subtilis*, *Micrococcus luteus*, *Staphylococcus aureus* and two Gram-negative strains *Pseudomonas aeruginosa*, *Escherichia coli*. The disc diffusion technique was followed at a concentration of 250 $\mu\text{g mL}^{-1}$ with Amacilin used as the standard drug. The zone of inhibition is compared using Table 3 and the corresponding clustered column chart is shown in Fig. 2. A good antibacterial activity has been possessed by all substituents in the microorganism in general. A satisfactory antibacterial activity against *B. subtilis*, *M. luteus*, *S. aureus* bacterial species when compared with standard Amacilin drugs. The substituents 3-Br, 4-F and 4-OCH₃ have moderate activity against *E. coli*. The substituents H, 4-Br, 3-Cl have excellent, 4-Cl and 4-NO₂ have very good activity against *P. aeruginosa*.

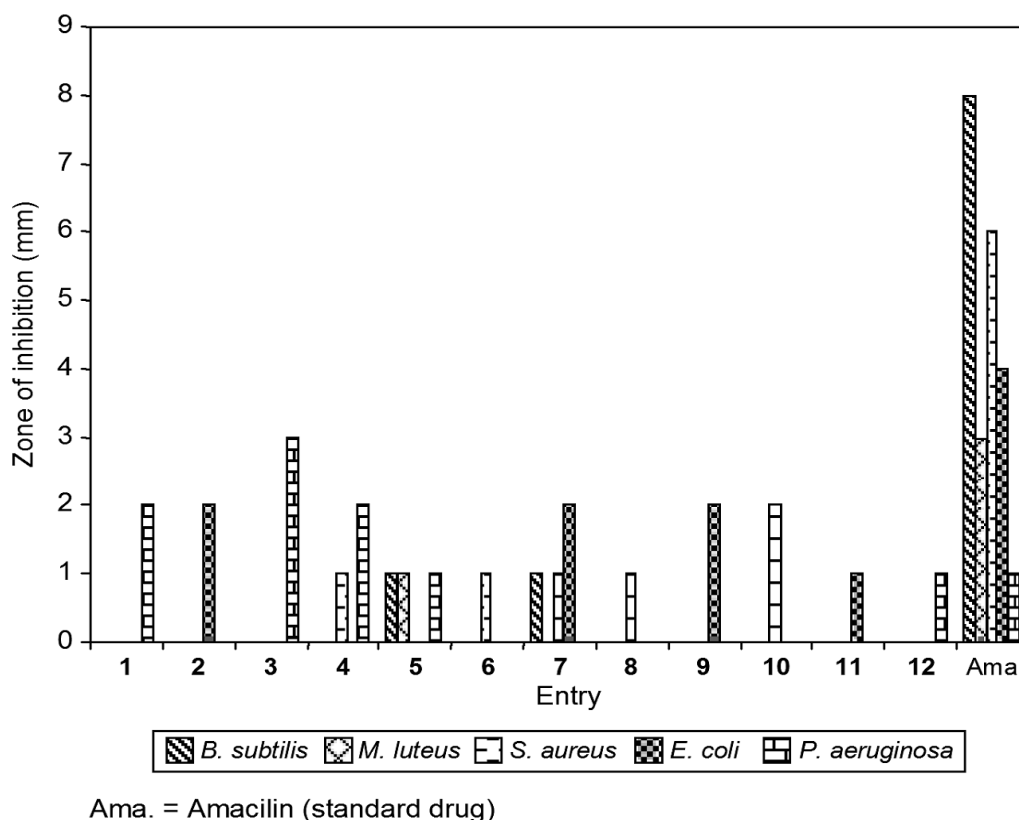


Fig. 2. Antibacterial activities of (*E*)-[4-(substituted benzylideneamino)phenyl](phenyl)methanones clustered column chart.

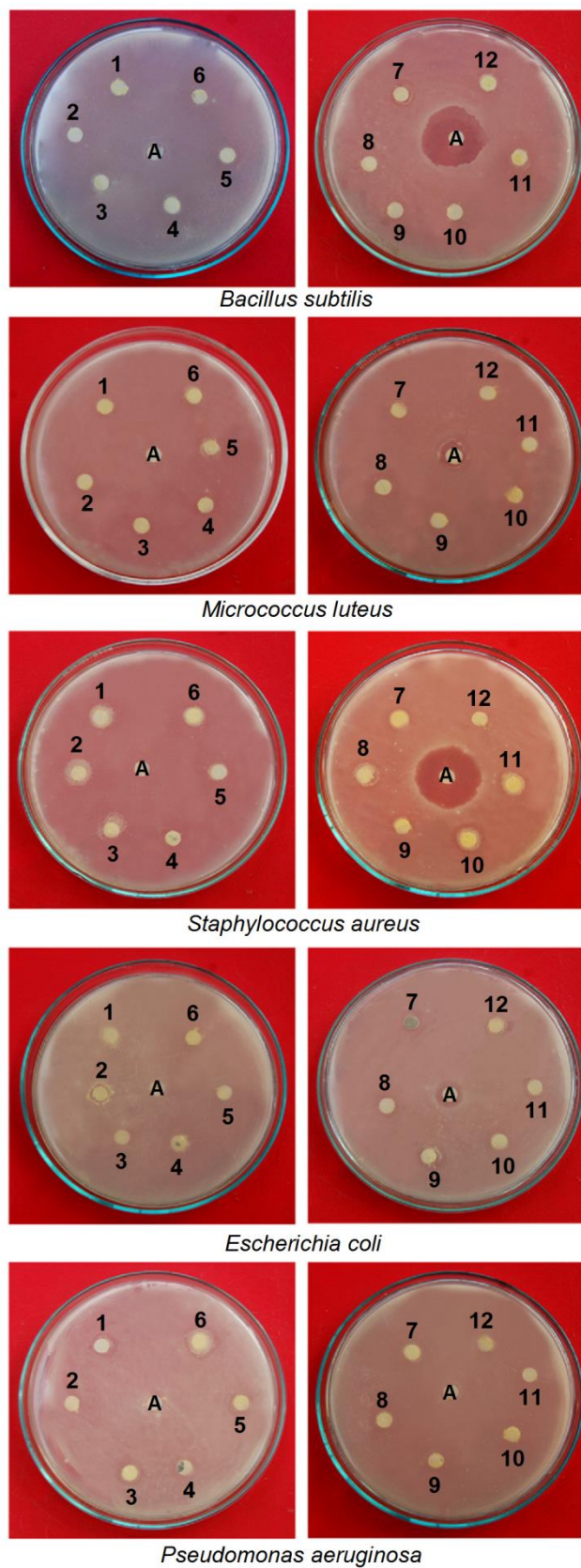


Fig. 3. Antibacterial activities of (E)-[4-(substituted benzylideneamino)phenyl](phenyl)methanones. A – Amacilin (standard drug).

3. 2. 2. Antifungal sensitivity assay

Antifungal sensitivity assay has been performed using disc diffusion technique [32] PDA medium has prepared and sterilized as above. It has been poured in the petri plate which has been already filled with 1 mL of the fungal species.

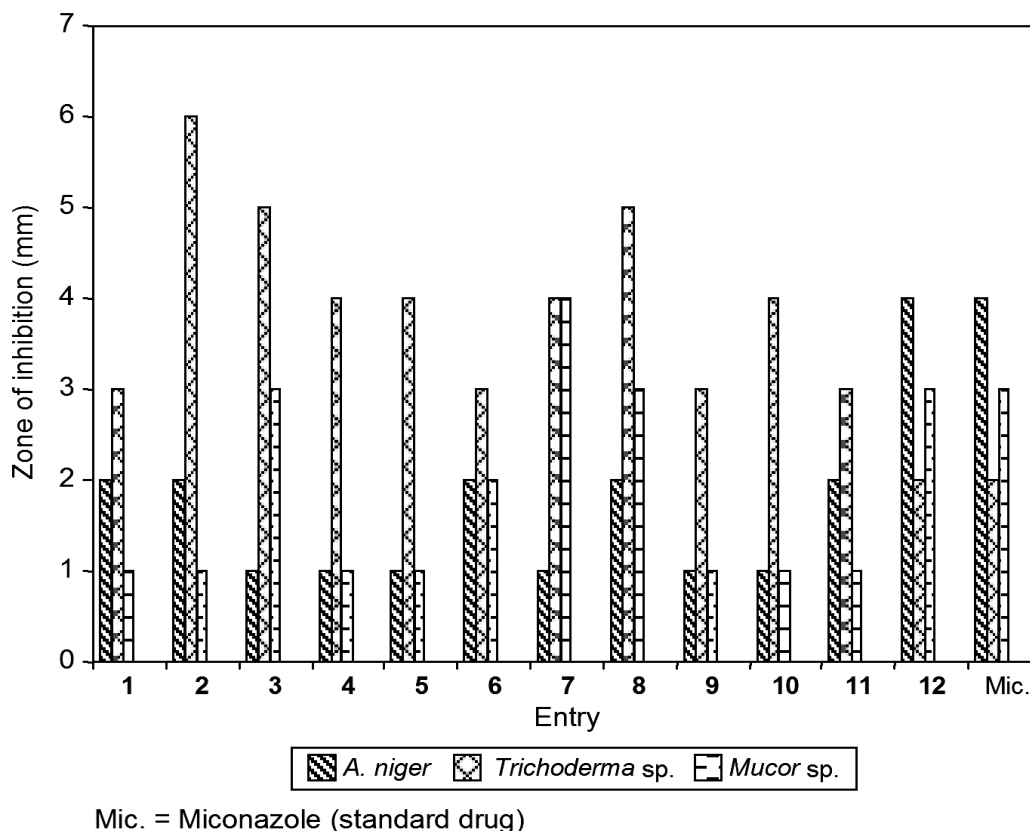


Fig. 4. Antifungal activities of (*E*)-[4-(substituted benzylideneamino)phenyl](phenyl)methanones clustered column chart.

The plates have been rotated clockwise and counter clockwise for uniform spreading of the species. The discs have been impregnated with the test solution. The test solution has been prepared by dissolving 15 mg of the Schiff's bases in 1 mL of DMSO solvent. The medium have been allowed to solidify and kept for 3 days.

Then the plates have been visually examined and the diameter values of zone of inhibition have been measured. Triplicate results have been recorded by repeating the same procedure. The antifungal activities of substituted imines have been studied and are shown in Fig. 5 and the zone of inhibition values of the effects is given in Table 4. The clustered column chart is shown in Fig. 4. A good antifungal activity has been possessed by all substituents on the microorganism in general. The substituents 4-NO₂ have excellent and H, 3-Br, 4N(CH₃)₂, 3-OCH₃ and 3-NO₂ have moderate activity against *Aspergillus niger*. The all substituents have been excellent activity against *Trichoderma* species. The substituent 4-F have excellent, 4-Br, 3-OCH₃, 4-NO₂ very good and 4N(CH₃)₂ have been moderate activity against *Mucor* species.

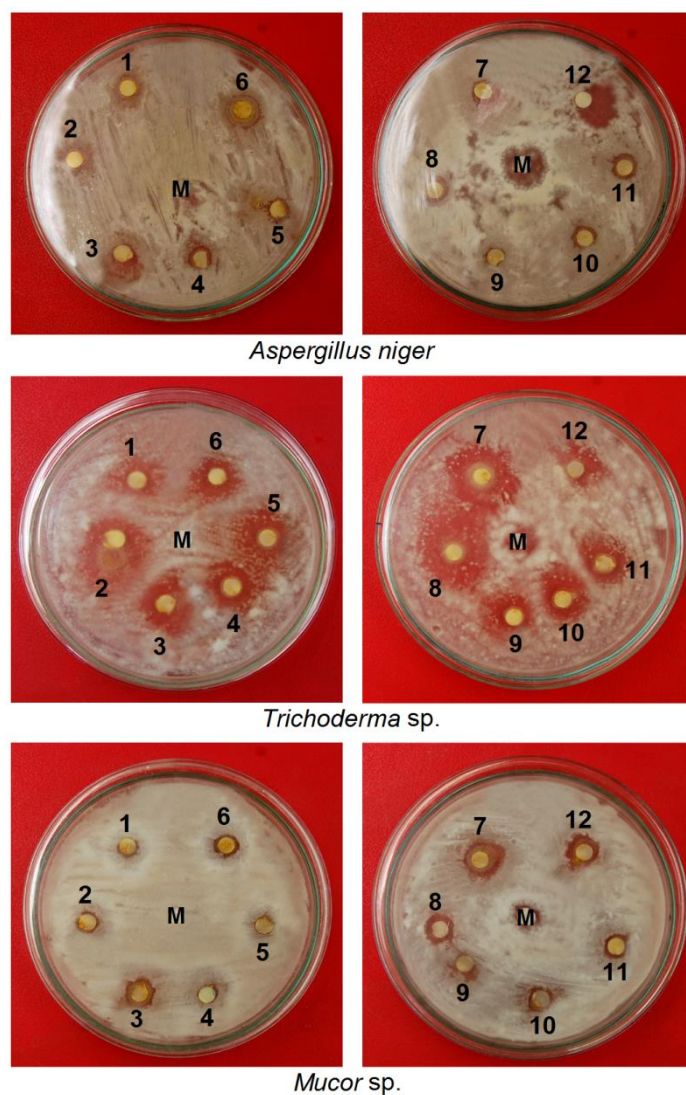


Fig. 5. Antifungal activities of (*E*)-[4-(substituted benzylideneamino)phenyl](phenyl)methanones. M – Miconazole (standard drug).

Table 4. Antifungal activity of (*E*)-[4-(substituted benzylideneamino)phenyl](phenyl)methanones.

Entry	X	Zone of inhibition (mm)		
		<i>A. niger</i>	<i>Trichoderma</i> sp.	<i>Mucor</i> sp.
1	H	2	3	1
2	3-Br	2	6	1
3	4-Br	1	5	3
4	3-Cl	1	4	1

5	4-Cl	1	4	1
6	4-N(CH ₃) ₂	2	3	2
7	4-F	1	4	4
8	3-OCH ₃	2	5	3
9	4-OCH ₃	1	3	1
10	2-CH ₃	1	4	1
11	3-NO ₂	2	3	1
12	4-NO ₂	4	2	3
Miconazole		4	2	3
DMSO		–	–	–

4. CONCLUSIONS

A series of Schiff's bases (*E*)-[4-(substituted benzylideneamino)phenyl](phenyl)methanone have been synthesized by condensation of arylamines and substituted benzaldehyde using microwave irradiation in the presence of fly-ash:PTS under solvents-free conditions. This reaction protocol offers a simple, eco-friendly, non-hazardous easier workup procedure and high yields. These imines were characterized by their physical constants spectral data. The IR, NMR spectral data of these imines have been correlated with Hammett substituent constants, F and R parameters. From the results of statistical analyses, the effects of substituents on the spectral data have been studied. The antibacterial activities of all synthesized imines have been studied using Bauer-Kirby method.

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