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Synthesis, Characterization and Antibacterial Activity of Schiff Base Hybrid from 2-Aminothiazole-pyrazolecarboxaldehyde

Rameshwar M. More[1], Archana B. Kadam[1], Sachin P. Dalvi[1], Vivek T. Humne[2], Subhash B. Junne[1,*]

[1]*P.G. Department of Chemistry, Yeshwant Mahavidyalala, Nanded – 431 602, Maharashtra, India.*
[2]*Department of Chemistry, Shri R.R. Lahoti Science College, Morshi – 444 905, Maharashtra, India.*

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ABSTRACT

The five Schiff bases hybrid have been synthesized from 2-aminothiazole and pyrazole-4-carboxaldehyde under basic condition. The structure of novel compounds were established on the basis of their elemental analyses IR, ^{1}H NMR and ^{13}C NMR, and then screened for their in vitro antimicrobial activity. Among them **3b** and **3d** showed excellent activity when compared to other derivatives. Other remaining derivatives showed moderate activity.

1. Introduction

The 2-aminothiazole is a significant scaffold containing nitrogen and sulphur atoms found in many drugs and having broad biological spectrum [1]. Various *N*-functionalized transformations in 2-aminothiazole is reported owing to its unique behavior towards diverse biological activities. The *N*-benzoyl substituted aminothiazole is evaluated for antitubercular and antibacterial activities [2], *N*-aryl-aminothiazole screened for the antibacterial and anti-inflammatory activities [3], hydrozone derivatives of 2-aminothiazole has been investigated for antifungal activities [4], urea and thiourea derivatives has been checked as anti-HIV agent [5,6]. To careful survey of the literature, it was found that the most significant and convenient formation of the Schiff base is from 2-aminothiazole and variety of aldehyde. Ding and his co-worker synthesized Schiff base from 2-aminothiazole with substituted benzaldehyde by using the mixture of piperidine-glacial acetic acid [7]. Singh *et. al.* reported the Schiff base from 2-aminothiazole with indole-3-aldehyde to checked the anti-inflammatory activity [8]. Besides, the complexes of different metals with Schiff bases has been reported in material chemistry and biological activities [9].

Over the past few decades, pyrazole scaffold has attracted a great attention due to its wide use in pharmacological activities [10]. Pyrazole derivatives are also widely used in materials science fields such as non-linear optics (NLO), optical limiting, electrochemical sensing, Langmuir films and photoinitiated polymerization [11-14]. Dhanya *et al.* has evaluated the Schiff bases of pyrazole-4-carboxaldehyde and trizole-*N*-amine for their in vivo antitumor activity against Ehrlich-ascites-carcinoma-(EAC-) bearing Swiss albino mice [15]. Khan and Poojary have independently synthesized series of Schiff base of pyrazole-4-carboxaldehyde and heterocyclic amine to screened their antibacterial activities, including stability of these Schiff base was checked by DFT method [16,17]. Schiff base bearing imidazole and pyrazole nucleus, showed potency against *E. coli* [18]. While, Schiff base derived from *N*-phenyl-pyrazole derivative with 2-aminophenol, reveled promising anticancer activity close to doxorubicin [19]. Owing to its ubiquitous use in drugs and natural product, none of reports have been found for such Schiff base of 2-aminothiazole and pyrazole in literature. Therefore, aforementioned to above significance and in continuation to our research towards for the synthesis of new heterocyclic hybrids and planned to synthesize the 2-aminothiozole-pyrazzolaldehyde Schiff base hybrid for antibacterial study.

2. Experimental Methods

2.1 Chemical Material and Apparatus

The ^{1}H NMR experiments were performed with 500 MHz spectrometer, and chemical shifts were expressed in ppm (δ) with TMS as an internal reference. J values were given in hertz. The ^{1}H and ^{13}C NMR spectra were referenced to the residual solvent signals (2.50 ppm for ^{1}H and 39.9 ppm for ^{13}C in DMSO-d6). ESI-TOF and quadruple mass analyzer types were used for the HRMS measurements. Column chromatography was performed on silica gel (100–200 mesh) in glass columns to purify the compounds and visualized with UV light (254 nm) and iodine as a strain. Commercially available reagents and solvents were purchase from Aldrich, Spectrochem, Research Lab Chemical and used without further purification.

2.2 Biological Materials and Apparatus

MIC was determined by performing serial dilutions. Compounds suspension was prepared by dissolving 0.1 g of the compound in 10 mL of DMSO respectively. This represents 10 mg/mL and was then serially diluted. Tube containing only growth medium and inoculated organism was considered as control without any compound in it. The tubes were incubated at 37 °C for 24 h. Subculture was done from each tube on nutrient agar (including the control) and incubated at 37 °C for 24 h. Four bacterial strains have chosen for the study namely *Escherichia coli* (*E. coli*), *Staphylococcus aureus* (*S. aureus*), *Pseudomonas* (*B. Subtilis*) and *Staph epidermidis*. The bacterial strains were maintained on Muller-Hinton (MH) agar medium.

*2.3 Chemistry: General Procedure for the Synthesis of Schiff Bases (**3a-e**):*

In a 25 mL round bottom flask, mixture of substituted phenyl-2-aminothiazole (0.61 mmol), pyrazole-4-carboxaldehyde (0.56 mmol), and piperidine (2.13 mmol) in toluene (10 mL) was stirred at 130 °C for 20 h. The progress of reaction was monitored by thin layer chromatography. After completion of reaction, mixture was evaporated under vacuum and then diethyl ether (1 mL) was added. The solid product was separated, Decant the solvent and the crude product was washed with diethyl ether (1 mL) and n-hexane (1 mL) to obtain pure Schiff bases (**3a-e**).

*Corresponding Author:sbjunne@gmail.com(Subhash. B. Junne)

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N-(4-phenylthiazol-2-yl)-1-(3-(4-chlorophenyl)-N-phenyl-azomethane (**3a**): Yellow solid; 109 mg; Yield 67%; mp: 141-149 °C; Rf: 0.7 (2:8:: EA:Petether); IR: 3030, 2982, 1610, 1530, 1450, 837; ^{1}H NMR (500 MHz, DMSO): δ 9.40 (s, 1H), 9.09 (s, 1H), 8.05 (d, J = 7.7 Hz, 2H), 8.01 (s, 1H), 7.96 (d, J = 7.2 Hz, 2H), 7.92 (d, J = 8.5 Hz, 2H), 7.64 (d, J = 8.5 Hz, 2H), 7.58 (t, J = 8.0 Hz, 2H), 7.48 – 7.41 (m, 4H), 7.36 (t, J = 7.3 Hz, 1H); ^{13}C NMR (126 MHz, DMSO): δ 172.6, 156.1, 152.8, 152.8, 139.1, 134.5, 134.3, 132.1, 131.0, 130.9, 130.1, 129.2, 129.2, 128.6, 128.1, 126.3, 119.6, 119.3, 113.5; Calcd. for $C_{25}H_{17}ClN_4S$: C, 68.10; H, 3.89; N, 12.71%; Found: C, 68.19; H, 3.90; N, 12.76%.

N-(4-(4-methoxyphenyl)thiazol-2-yl)-(1-(3-(4-chlorophenyl)-N-phenyl-pyrazol-4-yl)azomethane (**3b**): Yellow solid; 109 mg; Yield 54%; mp: 110-119 °C; Rf : 0.7 (2:8:: EA:Petether); IR: 3020, 2980, 1610, 1530, 1450, 825; ^{1}H NMR (500 MHz, DMSO) δ 9.40 (s, 1H), 9.08 (s, 1H), 7.90 (dd, J1 = 12.6, J2 = 9.2 Hz, 6H), 7.85 (s, 1H), 7.72 (d, J = 9.7 Hz, 2H), 7.64 (d, J = 8.5 Hz, 3H), 7.19 (d, J = 8.7 Hz, 2H), 3.80 (s, 3H); ^{13}C NMR (126 MHz, DMSO): δ 160.1.1, 156.0, 152.3, 132.1, 131.0, 130.8, 130.2, 129.7, 129.2, 129.0, 128.1, 127.7, 127.4, 127.3, 119.6, 119.3, 114.6, 113.8, 55.6; Calcd. for $C_{26}H_{19}ClN_4OS$: C, 66.31; H, 4.07; N, 11.90%; Found: C, 66.27; H, 4.56; N, 11.76%.

N-(4-(4-bromophenyl)thiazol-2-yl)-(1-(3-(4-chlorophenyl)-N-phenyl-pyrazol-4-yl)azomethane (**3c**): Yellow solid; 109 mg; Yield 70%; decomposed > 160 °C; Rf : 0.7 (2:8:: EA:Petether); IR: 3030, 2982, 1622, 1510, 1450, 856; ^{1}H NMR (500 MHz, DMSO): δ 9.40 (s, 1H), 9.08 (s, 1H), 8.09 (s, 1H), 8.05 (d, J = 7.7 Hz, 2H), 7.92 (dd, J = 8.5, 3.6 Hz, 4H), 7.66-7.58 (m, 7H); ^{13}C NMR (126 MHz, DMSO): δ 152.9, 151.6, 139.0, 134.4, 133.7, 132.3, 132.2, 131.8, 131.1, 131.0, 130.2, 129.3, 128.4, 128.1, 128.0, 121.7, 119.7, 119.2, 114.3; Calcd. for $C_{25}H_{16}BrClN_4S$: C, 57.76; H, 3.10; N, 10.78%; Found: C, 57.50; H, 3.67; N, 10.34%.

N-(4-(4-methylphenyl)thiazol-2-yl)-(1-(3-(4-chlorophenyl)-N-phenyl-pyrazol-4-yl)azomethane (**3d**): Yellow solid; 109 mg; Yield 58%; mp: 83-86 °C; Rf : 0.7 (2:8:: EA:Petether); IR: 3030, 2982, 1610, 1530, 1450, 837; ^{1}H NMR (500 MHz, DMSO) δ 9.40 (s, 1H), 9.08 (s, 1H), 7.92 (dd, J1 = 12.4, J2 = 9.1 Hz, 6H), 7.82 (s, 1H), 7.72 (d, J = 9.7 Hz, 2H), 7.61 (d, J = 8.5 Hz, 3H), 7.21 (d, J = 8.7 Hz, 2H), 1.7 (s, 3H); ^{13}C NMR (126 MHz, DMSO): δ 160.1.1, 156.0, 152.3, 132.1, 131.0, 130.8, 130.2, 129.7, 129.2, 129.0,128.1, 127.7, 127.4, 127.3, 119.6, 119.3, 114.6, 113.8, 22.1; Calcd. for $C_{26}H_{19}ClN_4S$: C, 68.64; H, 4.21; N, 12.31%; Found: C, 68.50; H, 4.54; N, 12.56%.

N-(4,5-dihydronaphtho[1,2-d]thiazol-2-yl)-(1-1-(3-(4-chlorophenyl)-N-phenyl-pyrazol-4-yl)-azomethane (**3e**): Brown solid; 109 mg; Yield 51%; decomposed > 130 °C; Rf : 0.7 (2:8:: EA:Petether); IR: 3057, 2952, 1605, 1520, 1450, 867; ^{1}H NMR (500 MHz, DMSO) δ 9.40 (s, 1H), 7.92 (dd, J1 = 12.4, J2 = 9.1 Hz, 6H), 7.82 (s, 1H), 7.72 (d, J = 9.7 Hz, 2H), 7.61 (d, J = 8.5 Hz, 3H), 7.21 (d, J = 8.7 Hz, 2H), 2.93 (t, J = 5.0 Hz, 2H), 2.76 (t, J = 5.0 Hz, 2H); ^{13}C NMR (126 MHz, DMSO): δ 160.1.1, 156.0, 155.1 152.3, 132.1, 131.0, 130.8, 130.2, 129.7, 129.2, 129.0, 128.1, 127.7, 127.4, 127.3, 120.6, 119.6, 119.3, 114.6, 113.8, 24.2, 23.5; ; Calcd. for $C_{27}H_{19}ClN_4S$: C, 69.44; H, 4.10; N, 12.00%; Found: C, 69.67; H, 4.34; N, 12.21.

3. Results and Discussion

The starting substrates, 2-aminothiazole and pyrazole-2-aldehyde are prepared according to literature methods [20,21]. Recently, we reported the synthesis of aminals from 2-aminothiazole and pyrazole-4-carboxaldehyde using combined catalytic amount of molecular iodine and acetic acid *via* the formation of Schiff base [22]. It was concluded that acidic condition was not favorable for the formation of Schiff base. Thereafter, we attempted basic condition to obtain aminothiozole-pyrazole-2-aldehyde Schiff base hybrid. We screened the different bases and solvents to optimize the reaction conditions (Table 1).

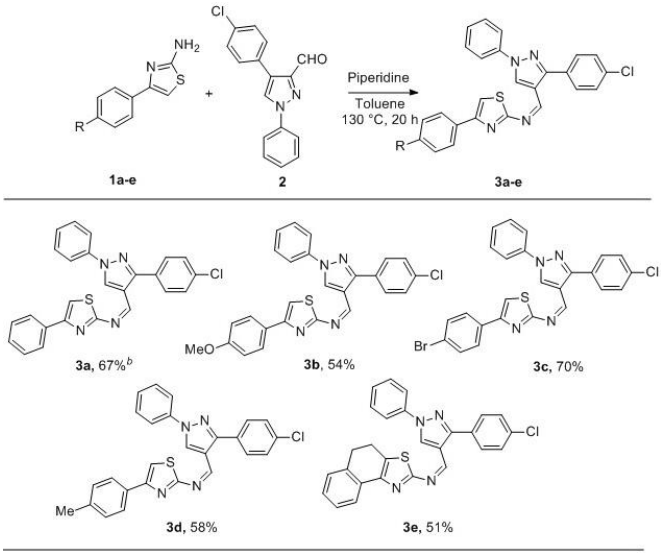
Table 1 Optimization condition

Entry	Base	Time (h)	Yield (%)[a]
a	Triethyl amine	24	36[b]
b	Piperidine	18	67
c	Morpholine	18	54
d	Pyrollidine	18	51
e	Potassium hydroxide	24	ND[c]
f	Sodium hydroxide	24	ND

[a]*Isolated product;* [b]*Starting substrate recovered;* [c]*No determined*

When piperidine used as a base, the desired product was achieved in good yield in 5 h. Other bases such as triethylamine, pyrolidine and di-isopropyl ethyl amine took longer reaction time with moderate yield. However, when sodium hydroxide and potassium hydroxide was added in the reaction mixture, TLC did not show presence of any starting substrates, while expected product could not be isolated from these complex reaction mixtures. Several solvents, such as ethanol, acetonitrile, dichloromethane, *N,N*-dimethylformamide, dimethylsulpoxide and toluene, were examined as the reaction medium. Toluene was found to be an efficient solvent for Schiff base formation. From the above results, it was concluded that a piperidine as a base and toluene as a solvent, were found to be a befitting condition for the formation of Schiff base from 2-aminothiozole and pyrazol-4-aldehyde.

With this optimal condition in our hand, five Schiff bases (**3a-e**) were synthesized. Gratifying, all the products were obtained in good yield. The prepared derivatives (**3a-e**) were then analyzed for structural confirmation by variety of analytical techniques such as FTIR, NMR, and elemental analysis (Scheme 1).



Scheme 1 Synthesis of pyrazole carbaldehyde and Schiff base derivatives[a]. [a]Reaction condition: **1** (0.61 mmol), **2** (0.56 mmol), piperidine (2.13 mmol) in toluene (10 mL) at 130 °C for 20 h; [b]Isolated yield

The disappearance of the band in the FTIR spectra of the compounds at 1718–1725 cm^{-1} and the appearance of the bands around 1609–1625 cm^{-1} due the formation of azomethine functional group. Further confirmation of the structures was carried out by the ^{1}H-NMR and ^{13}C-NMR spectra, the ^{1}H-NMR spectra exhibited the disappearance of the singlet around 10.032 ppm due to the aldehyde proton and simultaneous appearance of the singlet around 9.40 ppm due to the azomethine proton and ^{13}C-NMR spectra portrayed the disappearance of the signal around 186.8 ppm due to the HC=O carbon, and the appearance of the signal around 156.1 due to the azomethine carbon atom. Other signals were also observed around 114.24–116.01 ppm, 130.07–130.90 and 150.20–152.89 due to the three carbons of the pyrazole nucleus.

Table 2 Minimum inhibitory concentration (μg/mL) of pyrazole derivatives (1-15), Ciprofloxacin was used as positive control and negative control (DMSO) measured by dilution method

Compound	Minimum inhibitory concentration, μg/mL			
	Gram positive		Gram negative	
	S. aureus	*S. epidermidis*	*Pseudomonas*	*E. coli*
3a	50	100	100	50
3b	1.6	50	100	50
3c	12.5	100	100	100
3d	1.6	100	100	100
3e	3.12	50	100	Nil

Antimicrobial analysis of the compounds **3a-e** was performed against the microbes; *S. aureus*, *S. aureus*, *S. epidermidis*, *Pseudomonas*, and *E. coli* using dilution method to determination of minimum inhibitory concentration. The antimicrobial findings revealed that the compounds **3a-e** were found to exhibit very good activity against all microorganisms. While **3a-c** possess the moderate and compound **3e** was found less significant potential. The compounds **3d** having good antimicrobial activity against *S. aureus*. The detailed results of MIC reported in Table 2.

4. Conclusion

In summary, N-(4-aryl)thiazol-2-yl)-(1-(3-aryl)-N-phenyl-pyrazol-4-yl)azomethane Schiff bases derivatives were synthesized and characterized by IR, NMR, Mass spectroscopy and elemental analysis. Further synthesized compounds were evaluated for their antibacterial

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activities. The newly synthesized thiazole and pyrazole hybridized heterocyclic compounds exhibited moderate antibacterial activity against *S. epidermidis*, *Pseudomonas*, and *E. coli*, and significant antifungal activity against *S. aureus*. It can be concluded that these classes of compounds **3b** and **3d**. **3b** and **3d** classes of molecules have a good potential as active leads in medicinal chemistry. The further study to acquire more information concerning pharmacological activity is in progress.

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