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Article

A Short Review: Methodologies for the Synthesis of Schiff's Bases

Gayatri Phadnaik*

Department of chemistry, Institute of science, Nagpur, Maharashtra, India

ABSTRACT

Schiff bases are aldehydes or ketones like compounds in which carbonyl group is replaced by an azomethine or imine group. They exhibit a broad range of biological activities and are also used for industrial purposes. This short review compiles methodologies for the synthesis of Schiff bases and presents an overview of the use of Schiff base as a pharmacophore.

Key words: Schiff base, Azomethine, Pharmacophore, Biological activity, Green route.

1. INTRODUCTION

Condensation products of primary amines and carbonyl compounds are Schiff bases (also known as imine or azomethine). They were discovered by a German chemist, Nobel Prize winner, Hugo Schiff [1] in 1864. Organic compounds containing the azomethine (–HC=N-) group in their structure is called imines [2] (Figure 1).

Schiff base is a promising moiety in the area of synthesis and medical fields. The imine linkage has been found as an excellent bioactive and medicinally important moiety. Azomethines and their derivatives have been investigated [3,4] due to their striking complexometric behavior and pharmacological applications [5-9]. Due to these properties, it plays a major role in various biological activities [10-13] such as antibacterial [14-17], antiviral [18], antifungal [19-23], anticancer [24-27], anti-tubercular [28-30], anticonvulsant [31,32], anti-HIV [33], anti-helmintic [34], antifungal [35-38], anti-inflammatory [39], anti-nociceptive [40], anti-mouse hepatitis virus [41], inhibition of herpes simplex virus type-1 [42], adenovirus type-5 [43] anti-malarial [44,45], pesticidal [46], and herbicidal [47] activities. Schiff bases have the great potentials in different areas such as electrochemistry, bioinorganic catalysis [48], antioxidant property [49,50], metallic deactivators, separation processes, and in environmental chemistry [51].

2. BIOLOGICAL ACTIVITIES

2.1. Analgesic, Anti-inflammatory Activity

Sondhi *et al*. [52] reported the synthesis of N-(acridin-9-yl)-4 (benzo[d] imidazole/oxazol-2-yl) benzamides Schiff bases (Figure 2) which exhibit analgesic and anti-inflammatory activity. Bhandari *et al*. [53] derived it from 2-[(2,6-dichloroanilino) phenyl] acetic acid (Diclofenac acid) XVI and studied for their anti-inflammatory, analgesic, and ulcerogenic activities. Chinnasamy *et al*. [54] reported the synthesis of series of novel Schiff bases of Isatin. 3-(4-(4-Hydroxy-3- methoxylbenzylidene amino) phenyl amino) indoline-2-one exhibited better analgesic activity when compared to standard pentazocine. Bawa and Kumar [55] have synthesized Schiff base of 8-methyl-tetrazolo [1,5-a] Quinolineand evaluated their anti-inflammatory and antimicrobial activities. Lima *et al*. [56] have synthesized that [(4-dimethylamino benzylidene-3-(3, 4-methylenedioxyphenyl) propionylhydrazine] (Figure 3) was more potent than dipyrone and indomethacin are used as standard anti-inflammatory/antinociceptive drugs. Panneerselvam et al. [57] have been synthesized 4-(2-aminophenyl)-morph lines Schiff base and studied for their analgesic, anti-inflammatory, antibacterial, and antifungal activities.

2.2. Antimicrobial Activity

Raman *et al*. [58] synthesized a series of transition metal complex of Cu(II), Ni(II), Co(II), Mn(II), Zn(II), VO(IV), Hg(II), and Cd(II) from the Schiff base (L) derived from 4-aminoantipyrine, 3-hydroxy-4-nitrobenaldehyde, and o-phenylenediamine which have DNA cleavage activity and antimicrobial activity against *Salmonella Typhi*, *Staphylococcus aureus*, *Escherichia coli*, and *Bacillus subtilis* by the well diffusion method. Reddy [59] synthesized and investigated of new Schiff base and its solid metal complexes derived from p-Toluic hydrazide and 2-hydroxy-3-methoxy benzaldehyde(OVPTH) (Figure 4) using modified Sand Mayer's method. The compound has antimicrobial activity against *Salmonella* Typhi, *Enterococcus faecalis,* and *E. coli*.

2.3. Anti-tubercular Activity

Mamolo *et al*. [60] have synthesized [5-(pyridine-2- yl)-1, 3, 4-thidiazole-2-yl] acetic acid (3,4-diaryl-3H-thiazole-2-ylidene) hydrazide and tested for their *in vitro* antimycobacterial activity. Sinha *et al*. [61] have synthesized-arylidene- -[2-oxo-2-(4-arylpiperazin-yl) ethyl] hydrazide derivatives (Figure 5) from nicotinic acid and hydrazide hydrazones and evaluated their antimycobacterial activity.. Various diclofenac acid hydrazones (Figure 6) were synthesized and evaluated for their *in vitro* and *in vivo* antimycobacterial activities by Sriram *et al*. [62]. Hearn and Cynamon [63] have reported the synthesis of Schiff base the anti-tubercular activity of Schiff base.

2.4. Anticancer Activity/Antitumor

Demirbas *et al*. [64] have synthesized new hydrazide-hydrazones containing 5-oxo-[1,2,4]triazole ring (Figure 7) and studied their antitumor activity in breast cancer.

***Corresponding author:**
Email: gmphadnaik@gmail.com

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2.5. Anticonvulsant

Ragavendran *et al*. [65] synthesized that 4-Aminobutyric acid (GABA) is the principal inhibitory neurotransmitter in the mammalian brain. GABA hydrazones (Figure 8) were synthesized and evaluated for their anticonvulsant properties in different animal models Dimmock et al. [66] have synthesized acetyl hydrazones (Figure 9) provided good protection against convulsions while the oxamoylhydrazones were significantly less active.

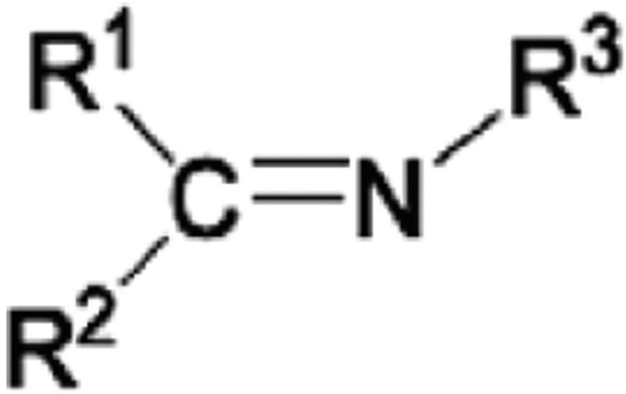

Figure 1: R^1,$R^2$and/ or R^3= alkyl or aryl.

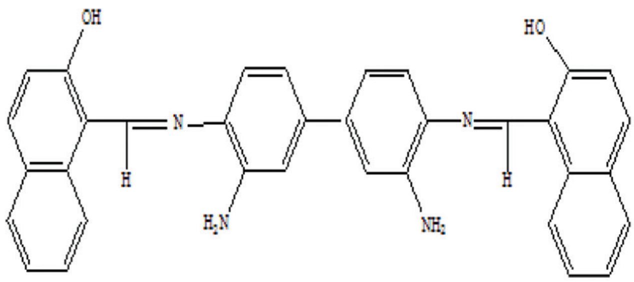

Figure 2: Anti-inflammatory agent

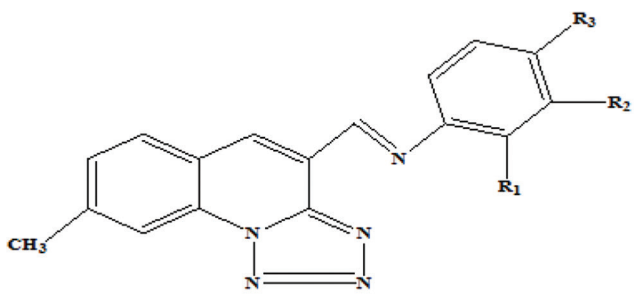

Figure 3: Antinociceptive agent

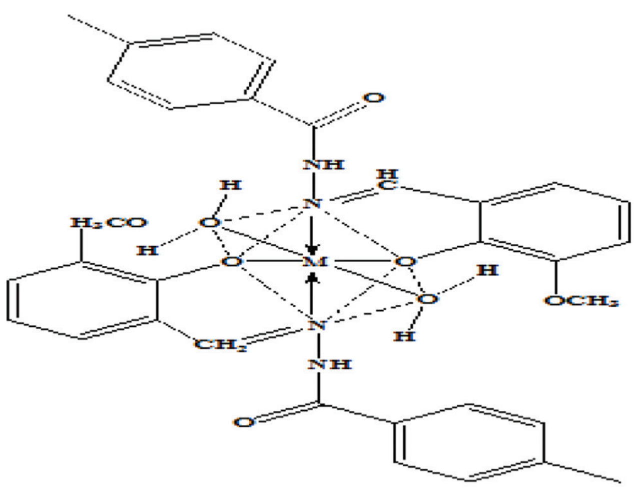

Figure 4: Antimicrobial agent

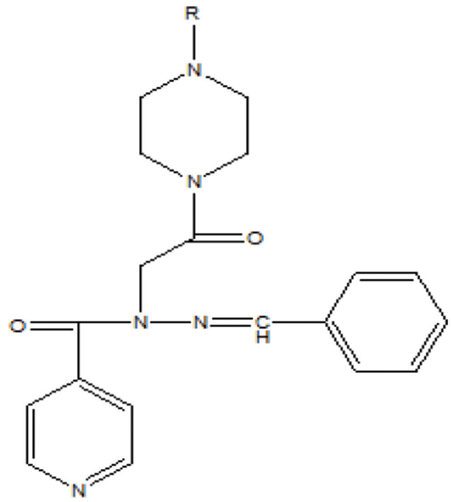

Figure 5: Hydrazide derivative

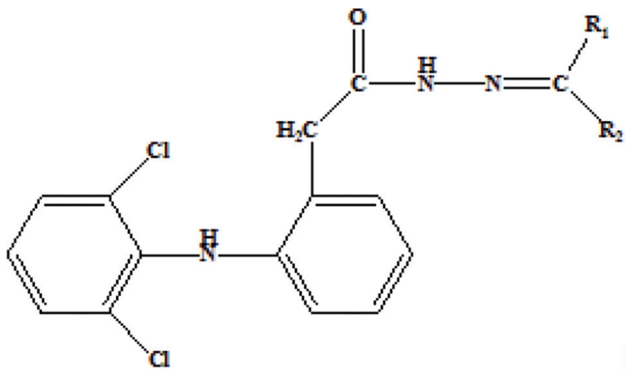

Figure 6: Antitubercular agent

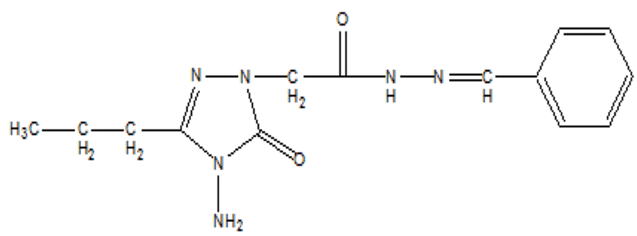

Figure 7: Antitumor agent

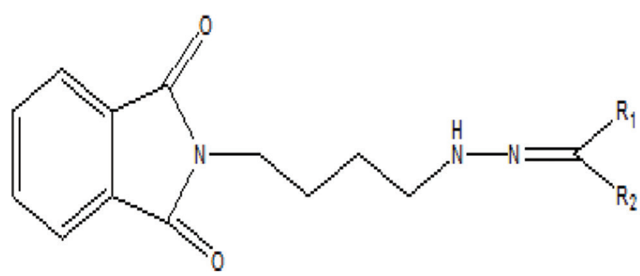

Figure 8: GABA hydrazones

Archana *et al*. [67] have synthesized new erindolylthiadiazoles and their thiazolidionones and formazans which have shown potential anti-convulsing activity.

2.6. Antioxidant Activity

Valentina *et al*. [68] have synthesized some substituted 1,2,4 - triazo-5-thione Schiff base and studied their antioxidant activity (Figure 10).

2.7. Antidiabetic Activity

A series of oxovanadium complexes with mixed ligands, a bidentate NN ligand, 37, and a tetradentate ONO-donor Schiff base ligand, (Figure 11) was synthesized and evaluated for protein tyrosine phosphate (PTP) inhibition. PTP1B has been identified as key enzyme related to insulin resistance [69].

2.8. Anti-hypertensive Activity

Shreenivas *et al*. [70] have reported many Schiff bases and they were prepared by condensation reaction of nitro compound containing biphenyl ether amines with aromatic aldehydes and ketone derivatives and thiazolidines were prepared by Schiff base with a thioglycolic acid. The synthesized compounds were screened for AT1 Angiotensin (An II) receptor antagonist activity. The nitro compound containing biphenyl ether Schiff bases and thiazolidines show good activity compared with losartan (Figure 12).

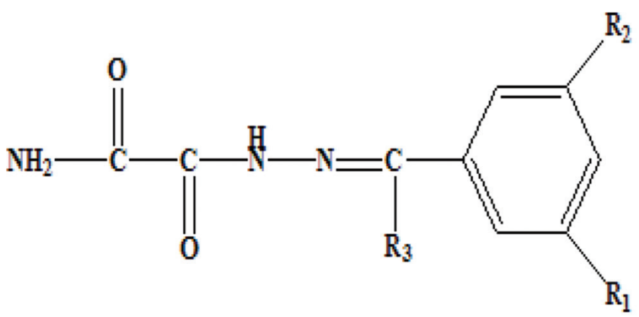

Figure 9: Anticonvulsant agent

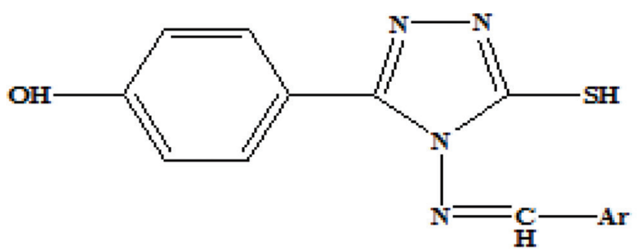

Figure 10: Substituted 1,2,4 - triazo-5-thione Schiff base

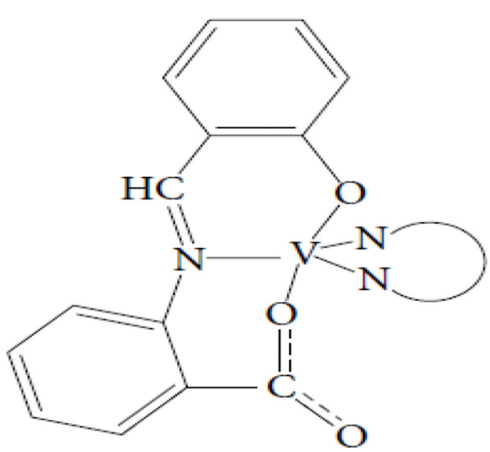

Figure 11: Tetradentate ONO-donor Schiff base ligand

3. METHODOLOGIES FOR THE SYNTHESIS OF SCHIFF BASES

The Schiff base is usually formed by condensation of an aldehyde or ketone with a primary amine according to the following scheme (Figure 13).

The mechanism of Schiff base formation is another variation on the theme of nucleophilic addition to the carbonyl group (Figure 14). In this case, the nucleophile is the amine. In the first part of the mechanism, the amine reacts with the aldehyde or ketone to give an unstable addition compound called carbinolamine. The carbinolamine loses water by either acid or base catalyzed pathways. Since the carbinolamine is an alcohol, it undergoes acid catalyzed dehydration.

3.1. Conventional Method

The Schiff bases are prepared (Figure 15) by refluxing a mixture of equimolar quantities of aromatic primary amine and substituted benzaldehyde in ethanol in the presence of 3–4 drops of glacial acetic acid.

3.2. Synthesis in Aqueous Medium

Green chemistry [71] has attained the status of a major scientific discipline. The studies of green chemistry have led to the development of cleaner and relatively benign chemical processes with many new technologies being developed each year. Among them, there is a large proportion of effort that has been devoted to the use of non-traditional solvent for chemical synthesis. Water is commonly considered as a benign solvent for its non-toxicity and abundant natural occurrence, water is undoubtedly the cleanest solvent on earth.

Leading methods for the synthesis of Schiff bases in aqueous medium are listed here with.

Tanaka and Shiraishia [72] carried out condensation reactions of aldehydes and amines occur efficiently in a water suspension medium, and the reaction products are collected easily by filtration.

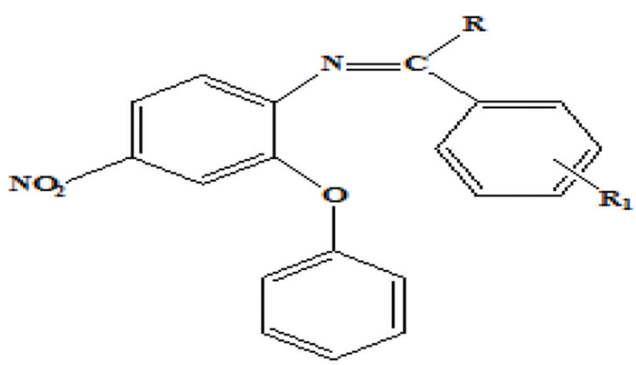

Figure 12: Biphenyl ether Schiff bases

Figure 13: Scheme I

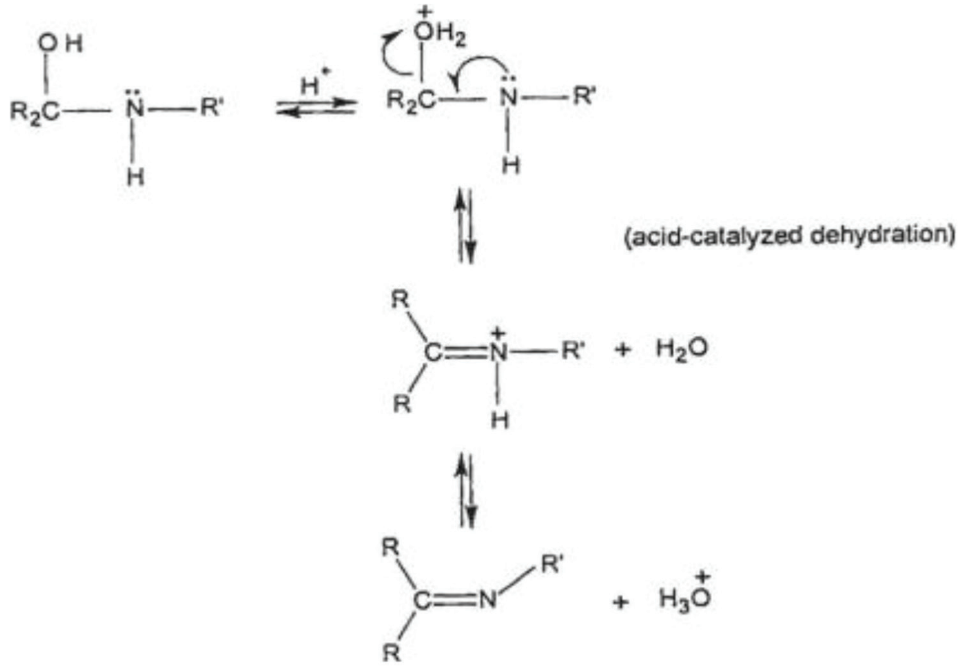

Figure 14: Scheme II

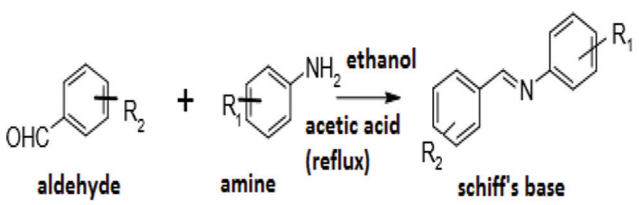

Figure 15: Scheme III

Singh *et al*. [73] gave an improved and facile synthesis of Schiff bases in aqueous medium.

Gupta *et al*. [74] performed water mediated condensation reaction of aldehydes and amines.

Rao *et al*. [75] (Figure 16) reported a novel and eco-friendly condensation reaction method permitting the "green synthesis" of various Schiff's bases by stirring 1,2-diaminobenzene with various aromatic aldehydes in water as solvent.

Zarei and Jarrahpour [76] synthesized pure azo Schiff bases (Figure 17) readily and conveniently in high yields by mixing of the reagents either as aqueous slurry, or by grinding at room temperature.

Murhekar and Khadsan [77] synthesized new Schiff bases by the condensation of 2-aminobenzothiazole with different aldehydes under organic solvent free condition efficiently in the presence of water.

Muskawar *et al*. [78] synthesized Schiff bases with polymer supported zinc–salen complex as highly efficient heterogeneous catalyst, in aqueous medium.

Thalla *et al*. [79] prepared that Schiff's base derivatives such as N'-(substituted- benzylidene)nicotinohydrazides (Figure 18) were synthesized by reacting nicotinohydrazide with various aryl/heterocyclic aldehydes containing pharmacological active functional groups under conventional conditions in ethanol as well as ultrasonic conditions in aqueous medium without using any catalyst.

Romanova *et al*. [80] synthesized Schiff bases from 3-amino-3-arylpropionic acid esters in aqueous medium.

Sachdeva *et al*. [81] carried green chemical one-pot multicomponent condensation reaction (Figure 19) of substituted 1H-indole-2,3-diones, various amino acids, and thiosemicarbazide is found to be catalyzed by lemon juice as natural acid using water as a green solvent to give the corresponding Schiff bases in good to excellent yields.

3.3. Synthesis by Microwave Irradiation [82-86]

Microwave irradiation has been used extensively to accelerate a variety of chemical reactions. Often, few minutes of microwave irradiation are sufficient for reactions that conventionally require several hours to reach completion. It appears that the increase in the frequency of molecular vibrations during microwave irradiation accelerates these reactions. It is also possible that microwave irradiation lowers the free energy of activation, thereby affecting the reaction rate. A thermal effect is another generally widely accepted mechanism through which allowed microwave irradiation can accelerate chemical reactions.

Green synthesis of salicylaldimine Schiff bases (Figure 20) was successfully carried out by irradiating salicylaldehyde with substituted aryl amines, respectively, without using any solvent and catalysts [87,88].

Some azomethines including substituted benzylidene-4-chlorobenzenamines (E-imines) have been synthesized by fly-ash: PTS catalyzed (Figure 21) microwave assisted condensation of 4-chloroaniline and substituted benzaldehydes under solvent-free conditions (SFC) by Suresh *et al*. [89].

Simple, rapid, clean, and environmentally friendly methods for the synthesis of Schiff bases and their cycloaddition with acetyl chloride to N-substituted phenyl-4-thiophenyl-2-azetidinones (Figure 22) under microwave irradiation are reported [90].

Khan *et al*. [91] synthesized that a series of pyrazole (Figure 23) containing Schiff [92] bases were synthesized, by the reaction of 3,5 dimethyl-1-phenylpyrazole-4-carboxaldehyde and the corresponding active amines under microwave irradiation.

Shinde *et al*. [93] (Figure 24) synthesized novel bis-Schiff bases from Propane-1,3-diamine on condensation with different halogen substituted benzaldehydes under microwave irradiation.

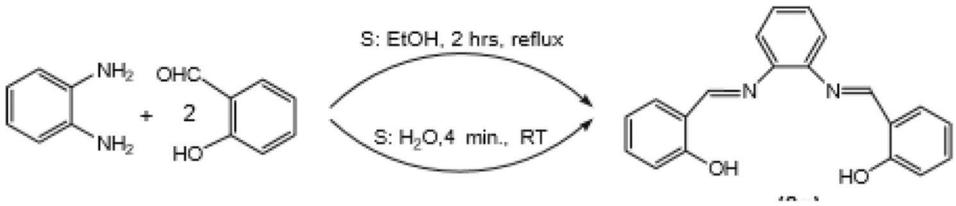

Figure 16: Scheme IV

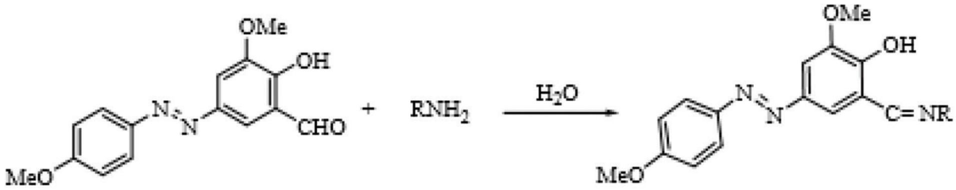

Figure 17: Scheme V

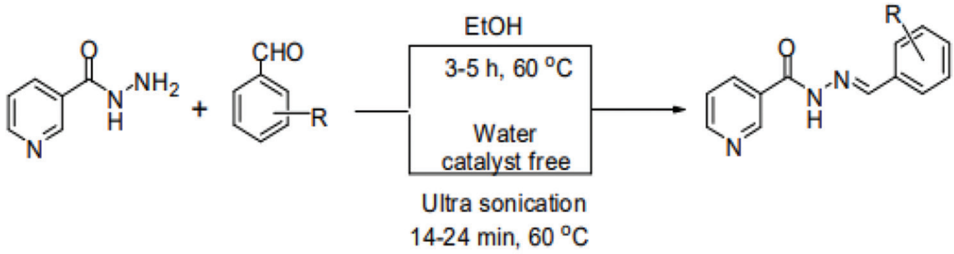

Figure 18: Scheme VI

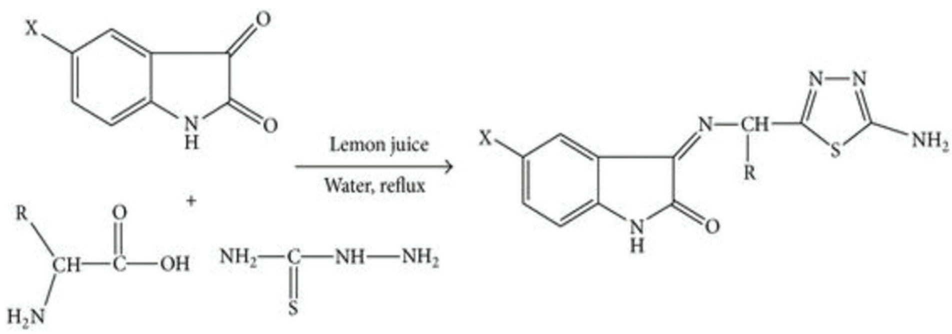

Figure 19: Scheme VII

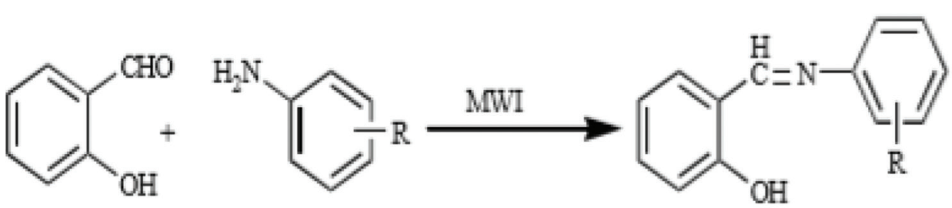

Figure 20: Scheme VIII

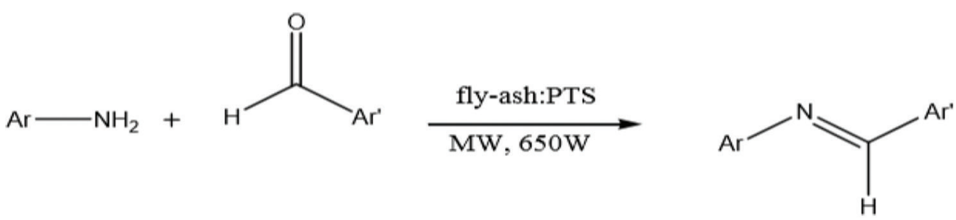

Figure 21: Scheme IX

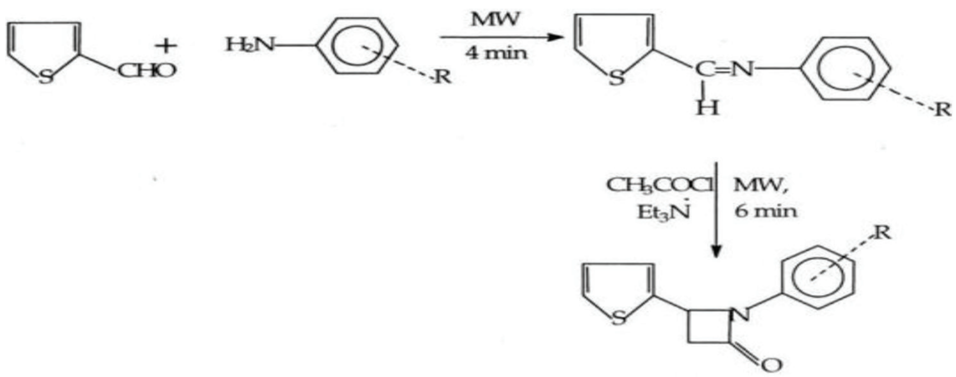

Figure 22: Scheme X

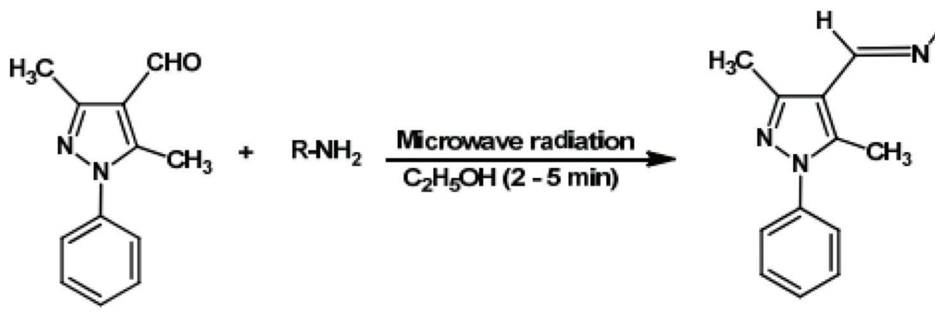

Figure 23: Scheme XI

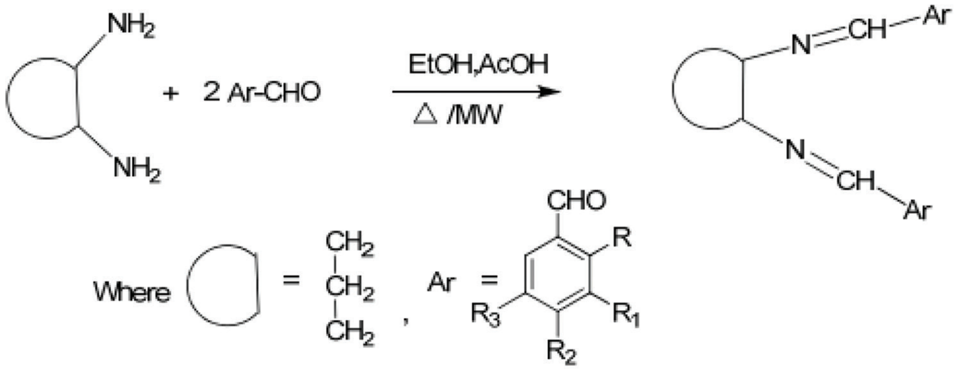

Figure 24: Scheme XII

Schiff base of Isatin [94] (Figure 25) was synthesized by condensation of the keto group of Isatin with different aromatic primary amines using microwave heating method.

New tetra Schiff bases were prepared in moderate yields through the condensation of different aromatic amines and bis-Schiff base in microwave synthesizer by Taha *et al*. [95].

Furthermore, new azo-Schiff bases were prepared by the condensation of with the azo-salicylaldehyde using the same method.

Desai and Desai [96] performed condensation of p-nitrobenzoyl hydrazide with substituted aromatic aldehydes under microwave irradiation.

A series of new Schiff's bases of Sulfanilamide were synthesized by Mohamed *et al*. [97] (Figure 26).

To optimize microwave assisted solvent-free synthesis of Schiff bases of substituted benzaldehydes and aromatic amines (3amino-6-bromo/Iodo-2-phenylquinazoline-4(3H) one) using wetting reagent ethoxyethanol (Figure 27). The goal of this study was to investigate

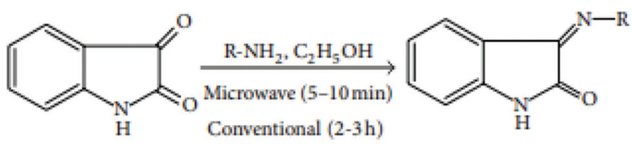

Figure 25: Scheme XIII

the % yields and time required for the completion of reaction for Schiff bases by microwave and conventional conditions [98].

Reaction of 6-methoxy-1, 3-benzothiazol-2-amine with substituted aldehydes under microwave irradiation [99] (Figure 28).

A series of compound 4-(2'-hydroxy-3'-chloro-5'-ethyl phen-1'-yl)-1-(4'-tolyl)-3-chloro-2-azetidinone have been prepared by the reaction of 2 -hydroxy-3 -chloro-5 -ethyl-N-(p-tolyl)-chalconimines with chloroacetyl chloride in the presence of triethylamine. The Schiff base derivatives have been prepared by the condensation of different substituted chalone derivatives with p-toluidine [100].

A fast and highly efficient method for the synthesis of some of the Schiff bases of amino thiazolyl bromo coumarin has been performed by microwave irradiation of 2′-amino-4′-(6-bromo-3-coumarinyl) thiazole and substituted aromatic aldehydes [101].

Rezaei *et al*. synthesized azomethines in high yields by reacting with hydroxylamine hydrochloride supported on melamine formaldehyde under microwave irradiation [102].

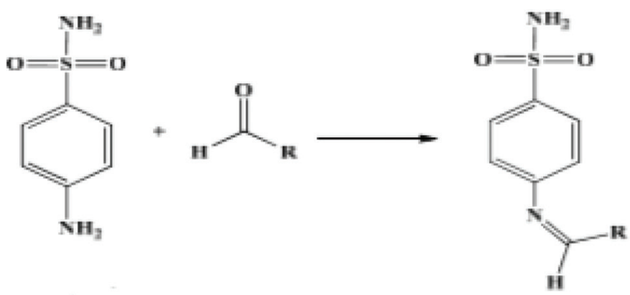

Figure 26: Scheme XIV

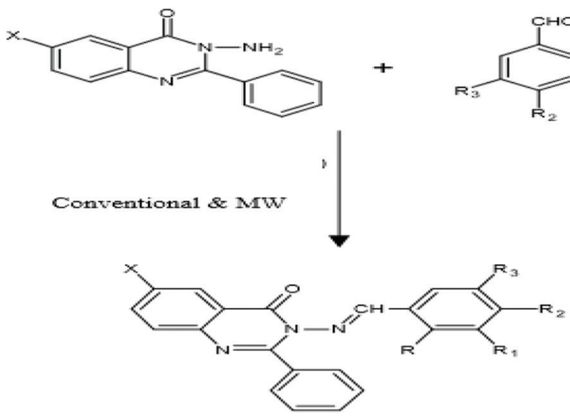

Figure 27: Scheme XV

Chakraborty *et al*., Yang and Sun and many performed comparative synthesis of Schiff bases by conventional and microwave method [103-105].

Two new Schiff base ligands containing −$SiOCH_3$ or −$SiOCH_2CH_3$ groups have been synthesized by the reaction of 2,4-dihydroxybenzaldehyde with 3-aminopropyltrimethoxysilane and 3-aminopropyltriethoxysilane. Six new transition metal Cu II, Ni II, and Co II complexes of these Schiff Base ligands were prepared by İspir *et al*. [106].

Microwave promoted synthesis of pharmacologically active Schiff bases of indole [2, 3-b] quinoxaline (Figure 29) was performed by Pai and Waghmode [107].

3.4. Synthesis in the Presence of Inorganic Salts

A simple and efficient method has been developed for the synthesis of some novel Schiff bases through the reaction of aromatic aldehydes with 2-aminobenzimidazole using catalytic amount of M(NO3)2 .xH2O (Figure 30) in an organic solvent at room temperature [108,109].

New Schiff base 2-[(4-Methyl-2-oxo-2H-chromen-7-yl)oxy]-N'-(substitutedmethylene) acetohydrazides were synthesized by the condensation of aryl/hetero aromatic aldehydes with 2-[(4-methyl-2-oxo-2H-chromen-7-yl)oxy] acetohydrazide using K_2CO_3 and $NaBiO_3$ (Figure 31) as catalyst [110].

An efficient green approach to the synthesis of Schiff bases [111] of 1-amino-2-aryl-3-oxo-1,2,4triazoles has been reported under $Mg(ClO_4)_2$ as catalyst, followed by the reaction with chloroacetyl chloride (Figure 32) in SFC to yield the azetidinones with excellent yields.

N-Sulfonyl aldimines [112] are powerful synthetic intermediates in organic synthesis and industrial application. They are prepared expeditiously under SFC by reaction between different aromatic aldehydes and sulfonamides in the presence of AlCl3 (Figure 33) in good to excellent yields.

P2O5/Al2O3 (Figure 34) is found to catalyze [113,114] the preparation of Schiff bases from the reaction of carbonyl compounds with primary amines efficiently under SFC.

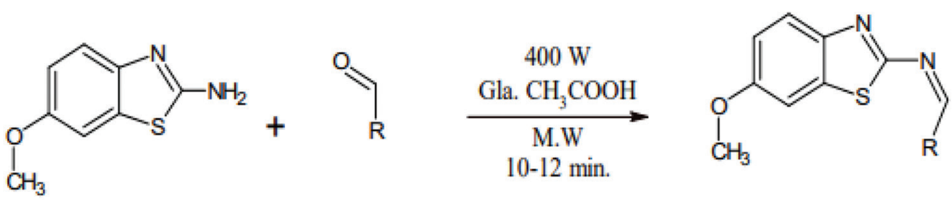

Figure 28: Scheme XVI

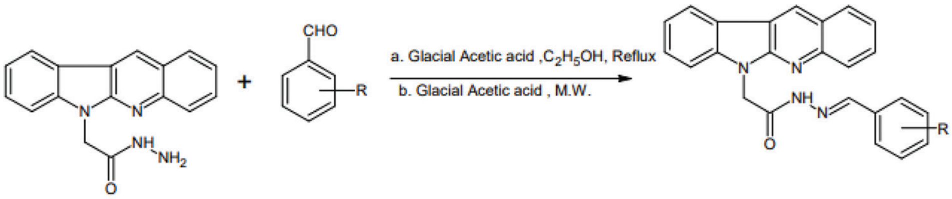

Figure 29: Scheme XVII

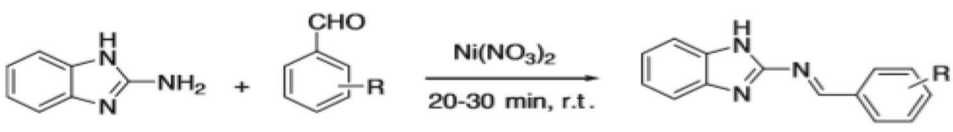

Figure 30: Scheme XVIII

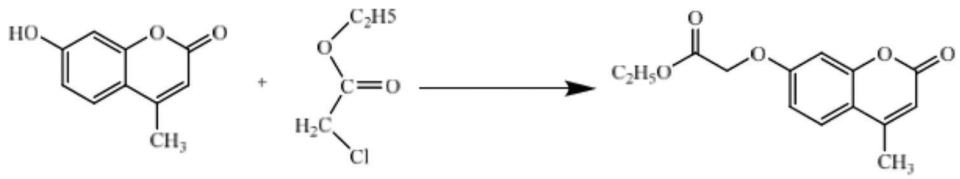

Figure 31: Scheme XIX

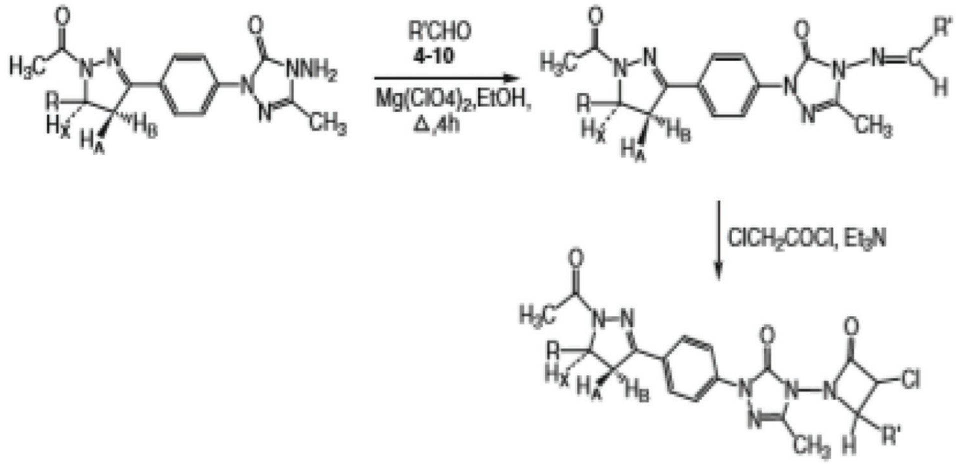

Figure 32: Scheme XX

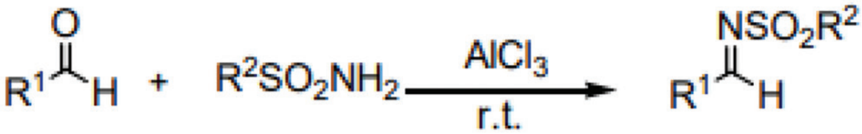

Figure 33: Scheme XXI

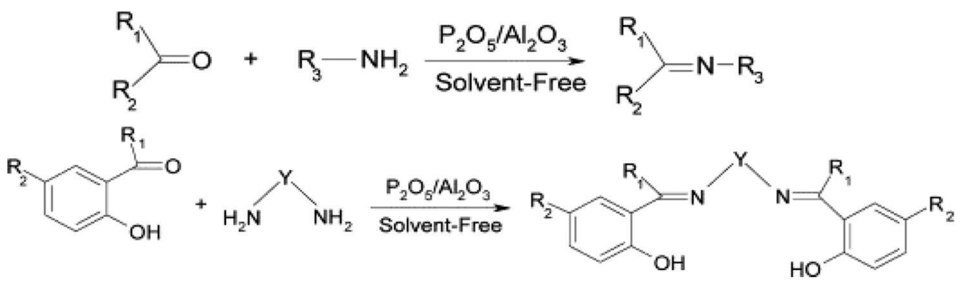

Figure 34: Scheme XXII

Vernekar *et al*. [115] and found that H3PO4.12WO3.xH2O catalyzes the preparation of Schiff bases from the reaction of 3-(1H-Benzimidazole-2-yl)naphthalene-2-amine with the different aldehydes efficiently in ethanol.

Siddiqui *et al*. [116] described the synthesis of Schiff bases by condensation of o-formyl phenoxy acetic acid and aryl aminothiazoles by reaction in hot ethanol or dioxane using sodium sulfate as a dehydrating agent.

The reaction of primary aromatic amines with aryl aldehydes is found to be catalyzed by cerium chloride heptahydrate under SFC to give the corresponding Schiff bases in good yields by Ravishankar *et al*. [117].

3.5. Green Synthesis

Green chemistry approach is an eco-friendly approach and has tremendous applications for the synthesis of various organic compounds and key intermediates in recent past. This technique involves an alternative reaction media to replace hazardous and expensive solvents routinely used in organic synthesis.

A series of amino Schiff bases have been prepared in good to excellent yield from the condensation of 1,2-diaminobenzenes with various aromatic aldehydes in presence of mango water [118] as natural acid catalyst under hand grinding technique (Figure 35).

A novel Schiff base as ON donor was synthesized by green methodology by condensation of 2-hydroxyacetophenone with furfurylamine (Figure 36) through microwave assisted reaction in fruit juice medium Sravanthi *et al* [119].

Yadav and Mani [120] described use of fruit juice of Citrus limetta, Vitislanata, and aqueous extract of *Mangifera indica* as natural acid catalysts for synthesizing Schiff bases, Wahab *et al*. [121] used natural acid found in natural products like tamarind and lemon.

Vibhute *et al*. [122] performed a facile and clean condition of 3,5-dichloro-2,4-dihydroxy benzaldehyde (Figure 37) to afford Schiff bases in quantitative yield using Grindstone [123] technique.

An efficient and eco-friendly synthesis of N1-(4-substitutedbenzylidene)-4-(tosylamino) benzo hydrazides (Figure 38) having sulfonamide pharmacophore have been carried in PEG-400 as greener medium at room temperature by Jagrut *et al*. [124].

β-Phenyl acrolein derivatives (Figure 39) have been successfully synthesized by Chigurupati *et al*. [125] and appear to be a novel and important class of antibacterial agents against Gram-positive and Gram-negative bacteria including *S. aureus*, *Pseudomonas aeruginosa*, and *Klebsiella pneumonia*.

4. SUMMARY

Schiff bases represent major pharmacophore with various biological properties, as some azomethine containing derivatives have already been used for therapeutic purposes. This literature review shows that Schiff base derivatives are pharmacologically very potent and, therefore, their design and synthesis is the potential area of research. It has been taken into account that the structural modifications of the basic structure of Schiff bases have allowed the preparation of new derivatives with a broad spectrum of biological activities.

This is an attempt to review the methodologies for synthesis of Schiff bases having potential biological activity.

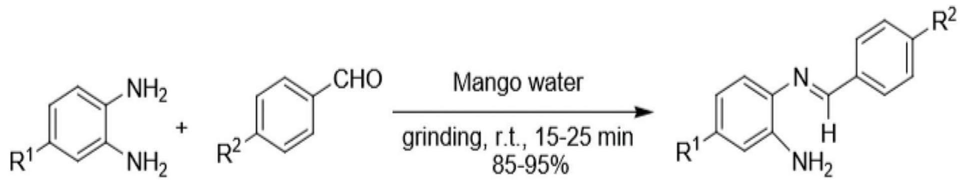

Figure 35: Scheme XXIII

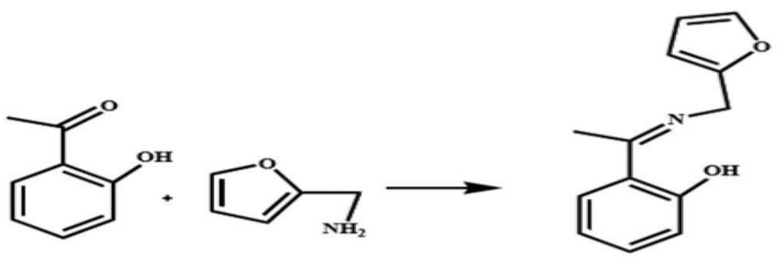

Figure 36: Scheme XXIV

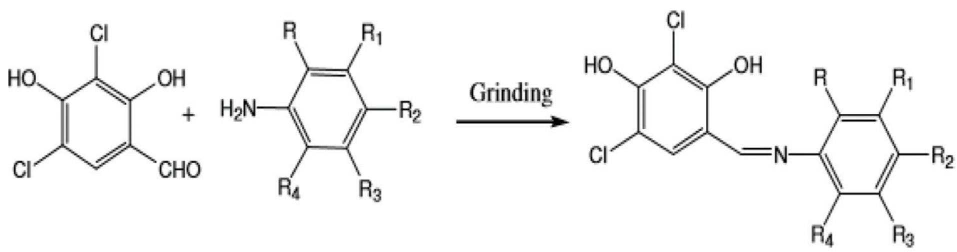

Figure 37: Scheme XXV

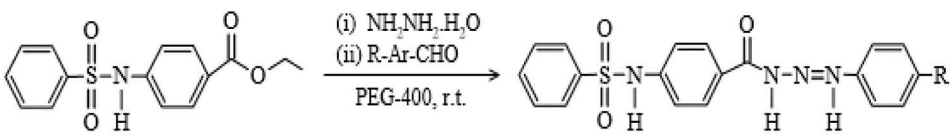

Figure 38: Scheme XXVI

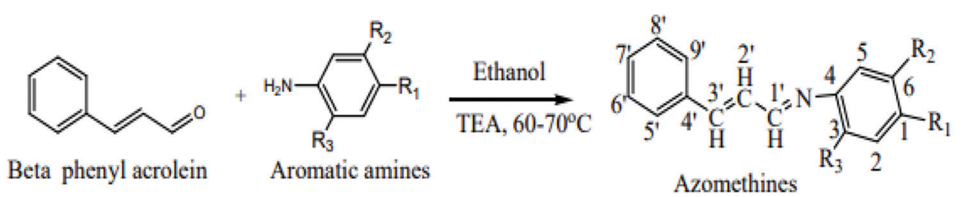

Figure 39: Scheme XXVII

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***Bibliographical Sketch**

Dr. Gayatri Mangesh Phadnaik, Associate Professor, Department of Chemistry, Institute of Science, Nagpur. Have eight (08) publications in the national and international journals of repute. Completed MRP of UGC in 2017. Attended National and international seminars and conferences for presenting research work.