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Chemistry

A REVIEW STUDY ON GREEN SYNTHESIS OF SCHIFF BASES

Ankit Sharma*	Department of Chemistry, S.P.C. Government College Ajmer (Rajasthan). *Corresponding Author
Dr. S. K. Arora	Department of Chemistry, S.P.C. Government College Ajmer (Rajasthan).

ABSTRACT Green chemistry offers significant financial and environmental advantages over traditional synthesis methods. It accomplishes this by subtly suggesting methods and tools. The present interest in green chemistry has proposed a further necessity for organic synthesis, where novel reaction conditions must locate, lowering the demand for hazardous ingredients or damaging organic solvents. These green approaches must improve selectivity, speed up reaction times, and facilitate product separation as compared to traditional routes. The first synthesis of Schiff bases was recorded by Hugo Schiff in the 19th century (imines). A primary amine and an aldehyde are often combined to form a Schiff base, which is an aldehyde with an azomethine group (C=N group) instead of a C=O group. In order to determine the best approach that gives greater yields in less time while maintaining an environmentally friendly atmosphere, this study aims to concentrate on the green synthetic techniques utilised for Schiff bases synthesis. The review considered a number of environmentally friendly synthetics methods, among which microwave irradiation is considered to be the best, followed by ultrasonic, employing natural acids, and grinding techniques.

KEYWORDS : Schiff base, green chemistry, green synthesis

Introduction:-

The term "green chemistry" refers to a contemporary area of study for organic synthesis and drug design techniques [1]. Microwave-induced amplification of organic processes is increasingly a topic of interest for chemists due to the quicker reaction durations, greater yields, and easier setup times compared to conventional techniques [2,3]. In microwave synthesis, low boiling, poisonous, and hazardous solvents are typically avoided to avoid accidents. Organic molecules may now be created using microwaves, which has been shown to be an efficient, safe, and environmentally beneficial process [4]. In compared to traditional methodologies, many of the technologies utilised by green chemists incorporate solvent-free reactions, simplify workup activities, cut reaction time, increase selectivity, and facilitate purification operations [5].

In the 19th century, Hugo Schiff reported the first synthesis of Schiff bases (imines). A Schiff base is regarded as an aldehyde that has an azomethine -N=CH- group in place of a C=O group. It is often created when primary amines and aldehydes react [8,9]. The chemical formula for Schiff bases is RCH=NR1 (where R & R1 denote alkyl or aryl substituents) [10, 11]. The synthesis of Schiff bases typically involves the nucleophilic addition of the NH_2 group to the C=O of the aldehyde, forming a hemiaminal compound in an azeotropic reflexing condition, and simultaneously removing water. The hemiaminal compound then dehydrates to produce an imine [12], as illustrated by the equation below [13]:

Applications of Schiff base:-

Schiff bases are generally acknowledged for being pharmacologically active compounds with a wide range of uses in the manufacturing of food [14], pigments and dyes, analytical and medicinal chemistry, as well as synthetic chemistry [7,9,15]. They serve as ligands for metal ion complexes and stabilisers in the production of polymers [15,16]. Many advantageous biological effects of schiff bases have been shown, including antituberculosis, antifungal, antimalarial, antibacterial, anthelmintic, anti-HIV, diuretic, anti-inflammatory, antiviral, antitumor, antiprotozoal, anticonvulsant, and analgesic properties [14,17-21]. The significance of Schiff bases is so quite apparent. By concentrating on green synthetic techniques for the synthesis of Schiff bases, this study aimed to determine the best way to produce higher yields in less time while preserving an ecologically friendly environment.

Schiff Base as a Corrosion Inhibitior:-

For industrial applications like pickling metals and steel, which often involves significant dissolving and metal consumption, severe acid solutions are frequently utilised. Corrosion inhibitors are frequently employed in these processes to regulate metal dissolution as well as consumption [22]. Metal corrosion is reported to be significantly inhibited by compounds with functional groups containing hetero-atoms that may donate lone pairs of electrons [23, 24].

In this respect, Schiff bases are well-known organic inhibitors of different metal corrosion in acid conditions because they at least include an azomethine-N [25,26]. The ability to readily and easily synthesis Schiff base compounds from very inexpensive material is their main benefit as corrosion inhibitors [25]. According to a study, Schiff bases' ability to block molecules is significantly higher than that of the comparable amines and aldehydes [27]. The Schiff bases' inhibitory efficiency is determined by the azomethine linkage, -HC=N-, which enhances the electron cloud in their backbone. This efficiency might vary depending on the kind of substituent that is present on the aromatic rings.

Conventional method for the synthesis of Schiff bases:-

In non-aqueous organic solvents, a combination of equimolar primary amines and aldehydes is typically refluxed for 8 to 12 hours to produce a schiff base. The reaction is often carried out using an azeotroping agent (if necessary) or by separating the generated water since it is acid catalysed [10,18,19]. Given that this reaction is very reversible, it is advantageous to remove the generated water from the process in order to maximise the yield of the Schiff bases [28,29].

Green synthetic methods for Schiff bases synthesis

1-Microwave irradiation method:-

Microwave-supported chemical processes have become a frequent technique in the synthesis of organic compounds. In comparison to traditional methods, this approach has several benefits, such as a considerably increased reaction rate that reduces reaction time, an increase in yield and product quality, and a simpler process [19]. Due to its eco-friendly characteristics, this process is also recognised as an important policy under "Green Chemistry". It has been demonstrated that using microwaves in organic synthesis results in quicker reaction times and more effective, risk-free, and environmentally friendly procedures [24, 25]. In the area of organic/pharmaceutical synthesis, drug firms are already using microwaves [26]. Because of their faster reaction times, cleaner settings, and ease of use, microwave aided processes (in solvent or solvent free conditions) have gained popularity. [27]

Bhusnure et al. 2015, synthesised schiff bases as potentially antibacterial compounds [19]. They employ 2-ethoxyethanol as a wetting agent and microwave irradiation as a heating source. The aldehydes and amines were placed in a beaker, and two drops of ß-ethoxyethanol were added. The ideal microwave settings were between 180 and 360W microwave power [19]. The reaction mixture was microwaved for around 3 minutes at (180W-600W) (with a brief cooling pause). Many Schiff bases may be produced using this straightforward microwave technique with short reaction times, excellent yields, and no generation of unwanted side products. They found that this strategy had an extra effect on the overall process compared to the traditional methods. Pooja et al. (2014) recognized a new set of Schiff bases by using glucose as a catalyst in the reaction of p-toluidine with various aromatic aldehydes in water. The setting for all of the reactions was a microwave [28]. Glucose has a lot of potential as an environmentally beneficial catalyst due to its total safety, biodegradability, low cost, and environmental friendliness. P-toluidine with 10 ml of an aldehyde and glucose solution combination. The reaction mixture irradiated MW at the specified time with (240 W). Items with high yields were bought [28]. By combining several substituted aldehydes with 4-aminoantipyrene, Seewan and colleagues (2018) created a collection of imines that were both dissolved in 100% ethanol. For one minute at 300W, the mixture was microwave irradiated. With 97% yields, they demonstrated the viability of this method of microwave-assisted synthesis [29].

In a recent study, Tapabashi et al. (2021) created a set of Schiff bases by irradiating the reaction mixture in a microwave reactor for 15 to 20 minutes at a moderate to high temperature after adding 3 to 4 drops of DMF. The same technique was followed but using 6–8 drops of glacial acetic acid in place of the DMF. Both techniques produce an acceptable to good output in a condensed amount of time [30].

2-Natural acid-catalyzed methods:-

The fact that natural acid catalysts (such as fruit juice) are an increasingly popular choice for chemical synthesis is largely due to their acidic nature, environmentally friendly attributes, enzymatic activity, and affordability. Focus is currently being placed on the value of fruit juice as a natural biocatalyst in organic synthesis [31]. At 25º C, the reaction is catalysed by the natural acid (for instance, lemon juice), which has a mild acidity. Juices from fruits and vegetables have recently been identified as potential organic solvents for the production of medicinal molecules [31].

Yadav and colleagues (2015) synthesised a number of Schiff bases using the stirring method with sweet lemon, grape, or unripe mango aqueous extract (as catalytic solvent). Equimolar volumes of benzaldehyde and aniline were mixed, then the natural acid catalyst (0.5–2.5 ml) was added, and the mixture was then preserved for 5–10 minutes [32]. The reaction mixture was also agitated for 2-4 minutes at 25° C. They discuss the protonation of heteroatoms that occurs during molecular conversion when natural acids are utilised. These natural juices block all of these organic juices, including malic acid, tartaric acid, citric acid, and oxalic acid. These acids give the required pH for the catalysed synthesis of Schiff bases [32].

For the environmentally friendly synthesis of nicotinic acid hydrazide Schiff base, Desai and Shinde (2015) developed a traditional method [33]. Nicotinic acid hydrazide was dissolved in ethanol, lemon juice was added while stirring, and after 15 minutes at room temperature, the aldehydes were added while stirring. They created a novel, simple, and ecologically friendly method for synthesising nicotinic acid hydrazide Schiff bases with high yields [33].

3- Ultrasonic methods:-

Thalla et al. (2012) developed a unique method to synthesize nicotinohydrazide Schiff bases without the requirement for a catalyst. By combining nicotinohydrazide and m-nitro benzaldehyde in 15 mL of water and subjecting the mixture to ultrasonic irradiation at 60 oC for 14 min, this novel synthesis technique produced 92% of the product as opposed to 84% by the traditional method [17]. They developed an efficient and ecologically friendly approach for synthesising Schiff bases, achieving the highest yields in an aqueous medium while using ultrasonic irradiation conditions and no catalyst [17].

Sonication is the process of using sound energy to excite particles for various purposes. A procedure known as ultrasonication commonly employs ultrasonic sound waves with a frequency of above 20 kHz [17, 34]. It is often used in the laboratory by using an ultrasonic bath or probe, also known as a sonicator [34].

In a different research, Nikpassand et al. (2013) developed an efficient, environmentally friendly, quick, and approachable method for the synthesis of azo-linked Schiff bases by the reaction of azo-linked aldehydes and aminopyrazole while being exposed to ultrasound [34]. Benefits of this approach include a reduced reaction time, moderate conditions, and greater yields. They combined azo-linked aldehydes and aminopyrazole in 10 mL of ethanol, and then they exposed the combination to 45 kHz ultrasound at 60 °C for 10 minutes in a water bath (5-15min). The yields of the pure products ranged from 85 to 96%. They concluded that the produced bubbles burst, which creates enhanced temperature and pressure and helps the intermolecular interaction, is what caused the greater yield and shorter reaction time by the ultrasonic approach [34].

4- Grinding method:-

Zarei and Jarrahpour created rapid, effective, environmentally friendly methods for the synthesis of Schiff bases in 2011 without the need of solvents or refluxing, with excellent yields and quick reactions [35]. To make the azo Schiff bases, the amine and aldehyde were mixed in a mortar and pestle for one minute before being heated to 25°C for 1.5 hours. At 70 °C, the generated water was extracted using a vacuum. The reactions evolved at room temperature with excellent yields in a rapid and inexpensive manner, and no base was necessary [35].

Sharma and Bhardwaj (2017) documented the solvent-free synthesis of Schiff bases using the amino acids glycine, phenylalanine, and tyrosine as well as salicylaldehyde [36]. The procedure uses a pestle and mortar for solid-state synthesis. No solvents are used at all during the process. Reactants are combined in equimolar amounts and then ground with a pestle for 10 minutes in a mortar. A yellow gum is created, and after 20 to 30 minutes of grinding continuously, it turns into a solid powder. This process is said to be straightforward and capable of producing a high yield. The molecular crystals' physical grinding action causes frictional heating, which drives the process forward [36].

Zarei et al. revealed the synthesis of novel Schiff bases in excess amounts by grinding at 25ºC [37] in Sachdeva and Khaturia's (2017) mini-review. A very excellent yield of azo Schiff bases was produced when azo-aldehyde and the appropriate amines were combined [38]. Meenakshisundaram and Manickam carried out the green technique, which involved grinding the reactants at room temperature and produced 90–95 percent (2019). Benefits of this unique technology include easy, safe, clean, and straightforward processing with excellent yields. To get the Schiff base (yield 70–75%) as solid, the dialdehyde and their respective amines were introduced to a mortar and ground for 10 min [39].

Hanadi (2020) uses a green method and the ball-milling approach to create novel Schiff base complexes. Additionally, Alharbia and his colleagues (2021) employed ball milling to get novel Schiff-base complexes from Fe, Ni, Zn, and Co ions, which is classified as a green process of synthesis [41].

5- Water as a green solvent

According to Rao et al., Schiff bases may be made in an aqueous medium with good yields that are higher than those of traditional methods (2010). Water is affordable, non-flammable, non-toxic, and safe to use, making it an ideal reaction media [1, 42]. So this approach has the benefit of not requiring any acid catalysts. It is crucial to note that water successfully drove the reactions [42], and the resulting Schiff bases may be separated by filtering. The response rate is accelerated by around 300 times utilising this method. Salicylaldehyde is added to a 1,2-diaminobenzene solution in 10 ml of water during the operation. The forming mixture was stirred for ten minutes at 25° C. When compared to the conventional method's 65% yield, the yield was 95%. For the synthesis of Schiff bases, this technique is regarded as an acceptable, simple, efficient, and green process [42].

Shamly synthesised Schiff bases based on salicylaldehydes in a different work (2018). In 10 ml of water, salicylaldehyde is combined with 3-amino benzoic acid, aniline, and ethylene diamine [42,43]. After 10 minutes of stirring at 25º C the combination produced an 83% yield. Water is used as a green solvent in this instance [43].

6- Comparison studies:-

Yang and his colleagues (2006) compare the microwave approach, the reflux method, and the use of anhydrous $MgSO_4$ [44] for the synthesis of simple Schiff bases. The yield was 85% when using the microwave approach, which involved placing a combination of p-toluidine, 3,4,5-trimethoxybenzaldehyde, and neutral alumina in DCM (2ml) in a microwave oven and irradiating it for 4 minutes at 20% output power. While the yield of the second procedure, which involved refluxing a solution of the identical reactants in 10 ml of benzene until no water produced (as measured by a Barrett distillation receiver), was 72%. In the third technique, anhydrous $MgSO_4$ was added to a stirred solution of the identical reactants in 10ml dichlorometham. The yield was 75%

after the resulting mixture swirled for 2 hours at 25°C. The microwave approach, in contrast, offers a significant benefit because it uses the least amount of time while producing the most [44].

Taj et al. (2011) produce Schiff bases under diverse conditions by using Lewis acids [46, 47] and sulphuric acid [45] as catalysts. The amines, aldehydes, and concentrated H_2SO_4 or glacial acetic (2 drops) are refluxed for 4 hours over water in the first method, which results in poor yields (30%). In step II, the same reactant was mixed with 5 ml of DCE and magnesium perchlorate $Mg(ClO_4)_2$, and the mixture was then refluxed in a water bath for intervals of three hours. Filtering was used to isolate $Mg(ClO_4)_2$. Using this strategy, excellent yields of 75% are generated [47].

In a different work, Arafa and Shaker (2016) used conventional techniques, sonication, microwave irradiation, and no catalyst to synthesis a collection of novel bis-Schiff bases [48]. Salicylaldehyde derivatives and diamines were suspended in ethanol in the traditional technique, and the mixture was then agitated for 10 to 12 hours. The same reactants were combined and microwave-irradiated for 6–10 minutes at 25°C using 450 W. The identical reactants were used in the ultrasonication procedure, but they were first dissolved in ethanol (5 mL), and then they were placed in an ultrasonic water bath at (25°C). followed by 1-4 minutes of ultrasonic exposure. By the time everything was said and done, they had seen that the ultrasonic approach had increased yield and decreased reaction rate. However, after increasing the irradiation periods, the yield (70–88%) from the microwave irradiation shows no appreciable improvement [48].

Kapadnis and his team (2016) examined four synthetic Schiff base preparation techniques [49]. The first microwave irradiation in which a few drops of 10% NaOH were given to the reactants to change the pH was performed. The reaction mixture was then exposed to radiation for 8 minutes in the microwave. In the second process, the prior mixture is refluxed for around 8 hours. The same prior mixture was agitated for five hours at room temperature using a magnetic stirrer as part of the third procedure. The last procedure was grinding the reactants with 10 ml of ethanol in a mortar and pestle, adding a few drops of citric acid to change the pH while it was being ground for around 20 minutes. The outcomes showed that the first approach had a significant benefit. It is ideal for industrial manufacturing, which has the best output and uses the least amount of time [49].

Several techniques for the synthesis of Schiff bases under microwave irradiation are mentioned in Shntaif and Rashid's (2016) overview research [50]. They either combined the reactants with or without alcohol, added acetic acid to the mixture together with ethanol, employed glacial acetic acid as a catalyst along with DMSO, or even used neutral alumina, silica gel, or concentrated sulfuric acid as catalysts. These processes were all carried out in a microwave environment. They reach the conclusion that these techniques were useful instruments for cutting down on reaction times and improving yields [50].

A comparison investigation employing several Schiff base synthesis techniques was conducted by Dayma et al. (2018) [51]. In the traditional procedure, a combination of salicylaldehyde and sulphanilic acid in ethanol was added, and the resultant liquid was agitated at room temperature for 4 hours. Using a magnetic stirrer at (25ºC) for one hour, the same reactants were dissolved in ethanol as in the room temperature procedure (few drops of acetic acid gradually added). In contrast, the reactants in the grindstone procedure were combined with a few drops of acetic acid and pounded in a mortar and pestle to create a yellow solid in 20 to 40 minutes. The ingredients for the microwave technique are dissolved in ethanol, one drop of glacial acetic acid is added, and the liquid is then microwaved at (140 W) for two to three minutes. The microwave approach produced the finest yields. The outcome also demonstrates the superiority of the grindstone approach over the conventional method. No organic solvent was required, and the reaction went quickly with acceptable yields [51].

Eftekhari et al research's on imidazole Schiff bases was undertaken in 2020 [52]. Heating, microwave irradiation, and the use of ethanol are all used throughout the synthesis. According to the data, it only takes 2-4 minutes to produce Schiff bases using ethanol techniques in maximum yields (90–98%). Microwave irradiation offers excellent advantages, such as cutting down reaction time from an hour to a minute and improving product yield [53].

Two Schiff bases generated from 4- aminobenzoic acid, 4-(4-(diethylamino)benzylidene)aminobenzoic acid, and 4-((4-methoxybenzylidene)aminobenzoic acid, were designed, synthesised, and chemo-photophysically characterised by Xochicale-Santana et al. in 2021. However, compared to conventional heating, the time and energy required to produce Schiff bases using green synthetic approaches (ultrasound and mechanosynthesis) is reduced [54].

CONCLUSION:-

Green chemistry offers significant financial and environmental advantages over traditional synthetic methods. Green synthesis techniques should make product purification simpler than traditional ones by improving selectivity, speeding up the reaction, and performing so rapidly. When primary amines and aldehydes interact, a Schiff base is synthesized. This study focused on the green synthetic methods employed during the synthesis of Schiff bases in order to identify the optimal strategy that produces higher yields in less time while maintaining an ecologically friendly environment. The microwave irradiation technique, which was found to be the best, was followed by the ultrasonic, employing natural acids, and grinding operations in the evaluation, which took into account a number of ecologically friendly synthetics technologies.

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