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IMIDAZOLE HETEROCYCLES: BIOLOGICAL ACTIVITIES, MECHANISM AND CLINICAL RELEVANCE.

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ABSTRACT

Imidazole-based heterocycles are a unique scaffold in medicinal chemistry because they may interact with a range of biological targets, coordinate metal ions, and generate hydrogen bonds. Imidazole derivatives are useful leads for drug development because they exhibit a wide range of biological actions, such as antibacterial, antifungal, antiviral, anticancer, anti-inflammatory, antidiabetic, and anti-HIV properties. Unique interactions are made possible by the five-membered ring with two nitrogen atoms. In order to facilitate binding to enzymes and receptors, one nitrogen functions as a hydrogen bond acceptor and the other as a donor or metal ligand. The relevance of emerging hybrids in contemporary pharmacotherapy is highlighted by their potential in clinical trials for targeted medicines, such as imidazole-quinoline conjugates for malaria and anticancer drugs overcoming drug resistance. SAR trends for imidazole derivatives in drug design center on changing the substituents of th e core five-membered ring to maximize potency, selectivity, and pharmacokinetics.

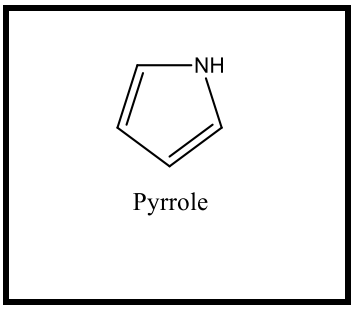
[Imidazole, Hydrogen bonding, Antimicrobial, Antifungal, Antiviral, Anticancer, Anti-inflammatory, Antidiabetic, and Anti-HIV actions, Synthesis, SAR]

INTRODUCTION

Heterocyclic compounds are predominantly of concern to the field of medicinal chemistry. Heterocyclic chemistry can be viewed as one of the most complex branches of all of chemistry. Synthetic heterocyclic chemistry has been important not only for its industrial and physiological significance, but also because of the diversity in its methods of synthesis and the theoretical implications associated with this type of chemical compound. Synthetic heterocyclic chemical compounds have been used in a variety of ways throughout all aspects

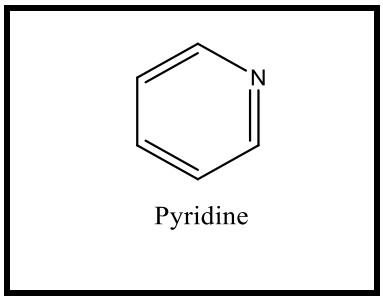
of our lives and have found application in agriculture, medicine, polymers and many other forms of industry. In general, the majority of synthetic heterocyclic chemical compounds serve as drugs that are available for use as: anticonvulsants, hypnotics, antineoplastic agents, antiseptics, antihistaminis, antivirals and anti-tumor agents. There are many new heterocyclic drugs introduced into the pharmacopeia each year. When looking at the size and the type of ring structure, combined with the effective substituent groups of the parent scaffold of a heterocyclic compound, the structure type(s) of the ring(s) can be shown to have a pronounced influence on their physicochemical properties.[1,2]

Imidazole, also known as 1,3-diazole, has a planar five-membered ring with two nitrogen atoms at positions 1 and 3 and three carbon atoms. Its chemical formula is $C_3H_4N_2$, and it is the most basic molecule in the imidazole series. Its nitrogen atom possesses a hydrogen atom similar to that of pyrrole-type nitrogen.



Pyrrole

Fig:1



Pyridine

Fig:2

It is soluble in water and other polar solvents due to its structure. It displays two equivalent tautomeric forms depending on which nitrogen atom carries the hydrogen. Imidazole's dipole moment of 3.61 D indicates that it is strongly polar and completely soluble in water. The 6π-electron system that gives it its aromatic property is made up of one electron from each of the other four ring atoms and the lone pair on the protonated nitrogen. Because of its amphoteric nature and acidic proton at N-1, it has a pKa of 14.5, making it slightly stronger than alcohols but less acidic than carboxylic acids, phenols, or imides. With a pKa of around 7 and a basic site at N-3, the conjugate acid is roughly 60 times more basic than pyridine in its basic function [3,4].

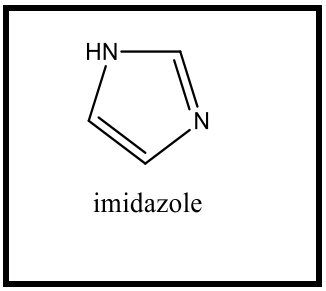


Fig:3

Physical Properties

Table: 1.

Property	**Value**
Appearance	White to Pale Yellow Crystalline Solid.
Odour	Slightly Amine- like
Melting Point	89-91ºC
Boiling Point	256-257ºC
Density	1.03g/cm^3(at 20ºC)
Solubility in Water	Miscible
Solubility in Organic Solvents	Soluble in Ethanol, Methanol, Acetone, DMSO, etc.

Chemistry of Imidazole

- Basicity: Because the nitrogen atom in the five-membered ring has a single pair of electrons that enable it to take a proton, imidazole acts as a weak base.
- Nucleophilicity: In a variety of processes, including nucleophilic substitution and addition reactions, the nitrogen atom in imidazole can function as a nucleophile.
- Coordination: Because imidazole contains a nitrogen atom, it may coordinate with metal ions to create metal complexes.
- Electrophilicity: In some reactions, especially electrophilic aromatic substitution reactions, imidazole can also function as an electrophile.
- Tautomerism: Imidazole displays tautomeric equilibrium between its 1H-imidazole and 3H-imidazole forms, in which the hydrogen atom's location on the nitrogen atom shifts.
- Oxidation: Under some circumstances, imidazole can undergo oxidation processes that produce imidazole derivatives [5,6].

History of Heterocyclic Chemistry

The history of the heterocyclic chemistry began in 1800s, in step with the improvement of organic chemistry. Some noteworthy developments-

- 1818: From uric acid, Brugnatelli isolates alloxan.
- 1832: Dobereiner produces furfural (a furan) by mixing starch with sulfuric acid.

- 1834: Runge isolates pyrrole ("fiery oil") by bones dry distillation.
- 1906: Friedlander discovered indigo dye, allowing synthetic chemistry methodologies to displace a large number of agricultural industries.
- 1936: Treibs syntheses chlorophyl derivatives from crude oil, explaining the biological source of petroleum.
- 1951: Chargaff's rules are explained, importance the role of heterocyclic compounds (pyrimidines and purines base) in the genetic code.[7]

General Synthesis of Imidazole

1. Radiszewski synthesis

Radiszewski states that when a dicarbonyl molecule, benzil, condenses with α-keto aldehyde, benzaldehyde, or α-diketones in the presence of ammonia, 2, 4, 5-triphenyl-imidazole is created. The usual Radiszewski Synthesis reaction.[8]

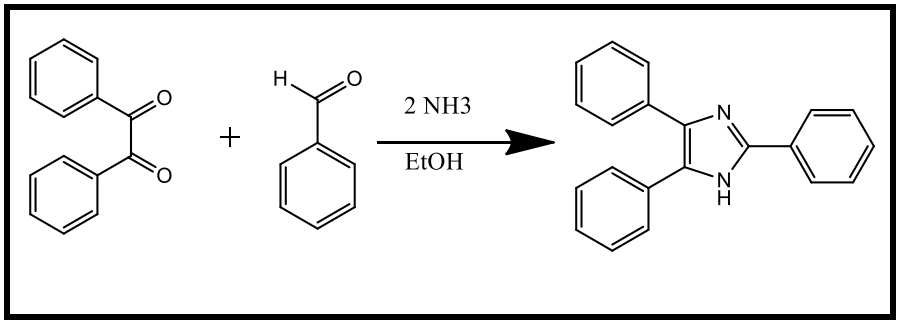


Fig:4

2. Imidazoline dehydrogenation

The gentler reagent barium magnate may convert imidazolines into imidazole when sulfur is present. Imidazolines generated from 1, 2 ethane diamine and alkyl nitriles react with BaMnO4 to form 2-substituted imidazoles.[8]

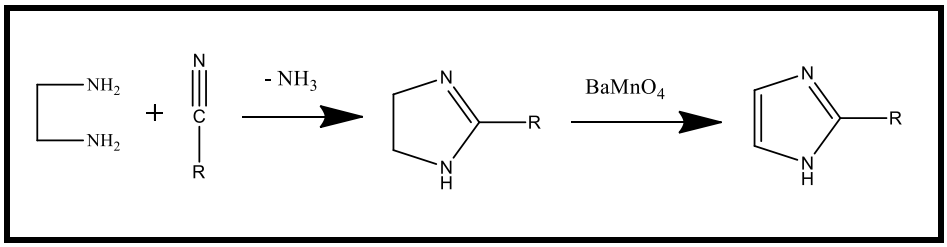


Fig:5

3. Wallach Synthesis

Wallach claims that treating N, N-dimethylox amide with phosphorus pentachloride produces a chlorine-containing molecule that, when reduced with hydroiodic acid, generates N-methyl imidazole. Under the same circumstances, N-diethylox amide is converted to a chlorine molecule, which then reduces to 1-ethyl–2-methyl imidazole.[8]

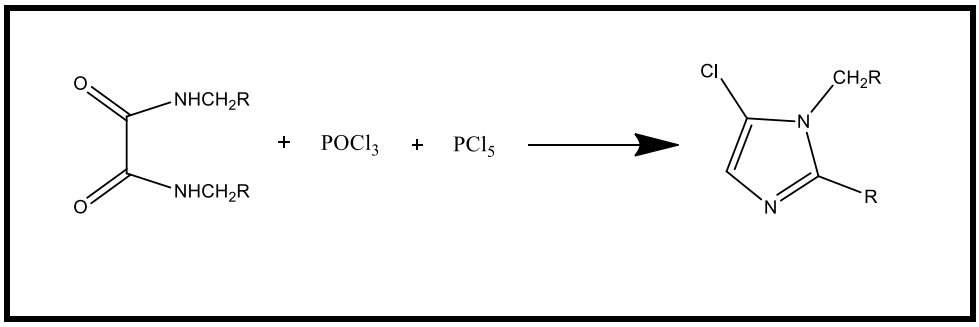


Fig:6

4. α-Halo Ketone

This method depends on how imidine interacts with alpha halo ketones. This method has been successfully used to synthesize 2, 4- or 2, 5-biphenyl imidazole. Similar to this, acyloin reacts with amidine or alpha halo ketones to form imidazole.[6,9]

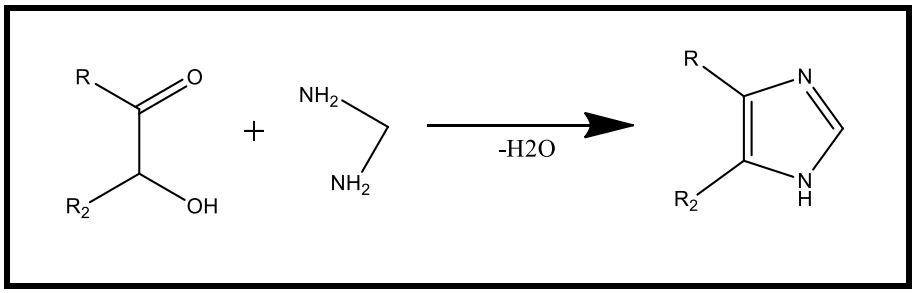


Fig:7

5. Synthesis of Markwald

Using α-amino ketones or aldehyde and potassium thiocyanate of imidazole with a 2-thiol substitution, 2-mercaptoimidazoles are produced. Using a variety of oxidative techniques, the sulfur may be readily removed to provide the required imidazole. [6,10]

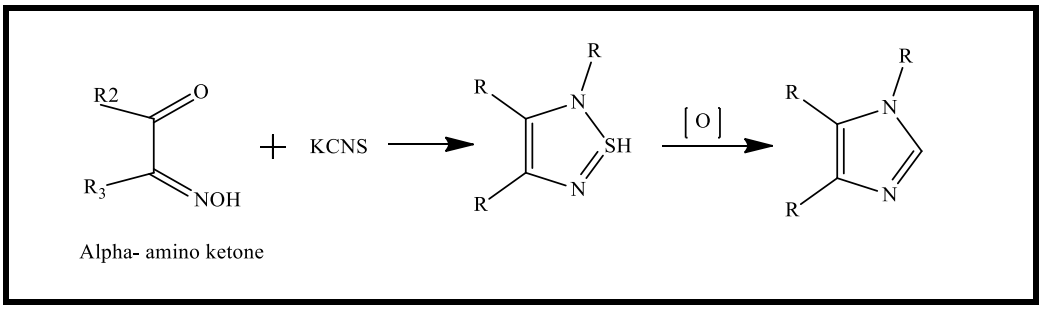


Fig:8

6. From hydroxyamino ketone, ammonia, and aldehyde

Hydroxyamino ketone reacts with ammonia and aldehyde to form substituted imidazole.[6,11]

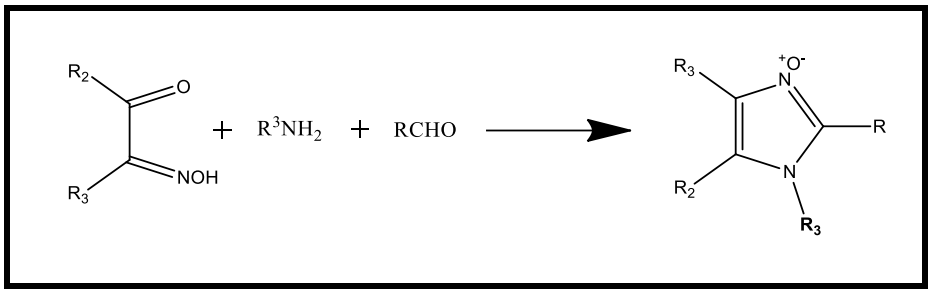


Fig:9

1. From aminonitrile and orthoformate

Substituted imidazole was created when a combination of aminonitrile and orthoformate condensed under the proper reaction conditions in the presence of a primary amine, as shown below.[6,12]

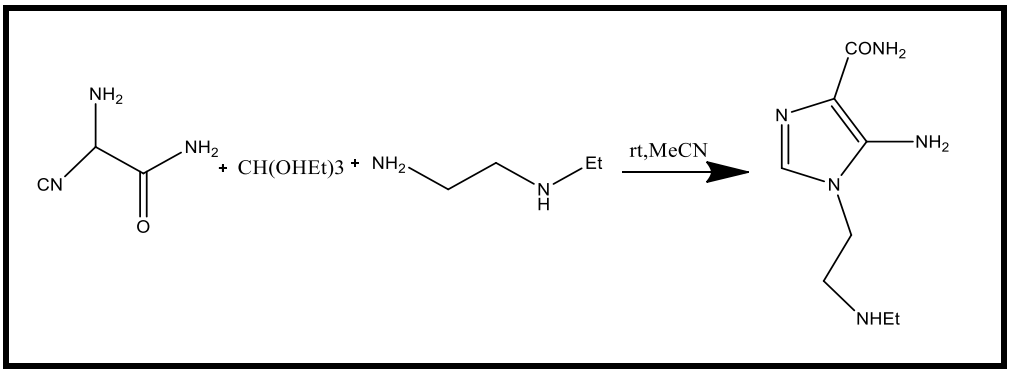


Fig:10

8.From formaldehyde and tartaric acid dinitrate

The simplest method for producing imidazole on its own is to heat the dicarboxylic acid in quinoline when cooper is present after ammonia reacts with formaldehyde and tartaric acid dinitrate.[6,13]

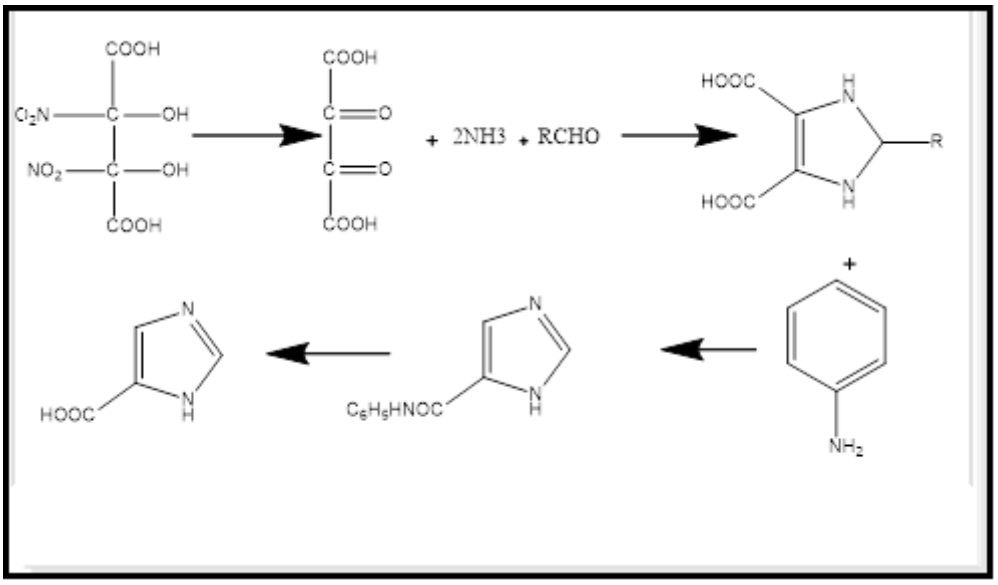


Fig.11

9. Cyclization of N-halo amidines with sodium ethoxide

N-haloamides are cycled with sodium ethoxide through a nitrene intermediate to produce benzimidazoles.

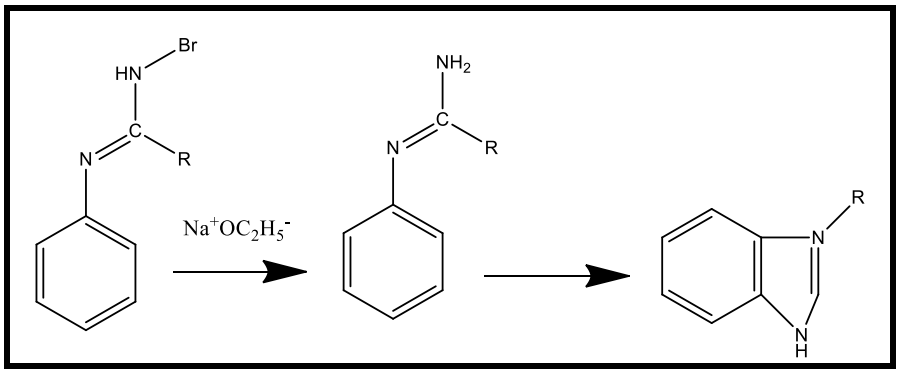


Fig:12

Imidazole Ring Structure Activity Relationship (SAR)

Imidazole compounds exhibit a variety of biological effects, and structural alterations are associated with their effectiveness. The compound's capacity to bind to certain receptors is impacted by substitution at position 2 of the imidazole ring. The molecule's electrical and

lipophilicity characteristics are changed at positions 4 and 5. Optimizing metabolic pharmacokinetics requires modifying the nitrogen atom (N-1).[14]

One way scientific properties of a molecule such as Electronics affects on how the will work in biological systems. For example, how much electron density is distributed within the chemical ring creates the value of interaction of these types of compounds will have to biochemistry.Placing a group that withdraws an electron at the 2-position of the imidazole ring has been shown to substantially increase antimicrobial properties, while a substituent that adds electrons at the same position produces an increase in anti-inflammatory properties.[15]

The three-dimensional arrangement of the functional groups on the imidazole ring, such as the ability of these groups to exist in either a left handed or right handed arrangement, is critical to the specific binding affinity of the compound to its parent site, but in other words, how well the inhibitor will bind to the enzyme or protein.[16]

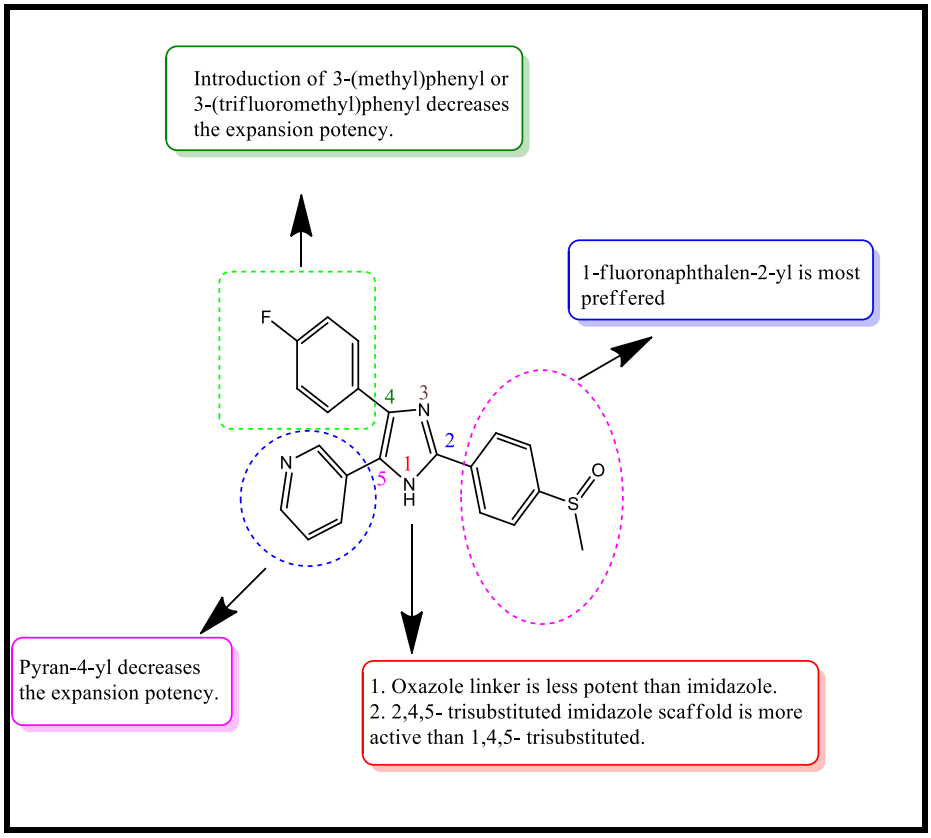


Fig:13

- **N-1 Location Substitution**- The substitution of the nitrogen at position 1 has significant importance for the biological activity of the molecule, with both N-1 and N 3 methyl results in a loss of bioactivity.

- **Lipophilic Modifications**- Adding lipophilic groups, especially five or six membered rings, increases the biological activity of this compound.
- **Aromatic Ring Substitution at Positions 2, 4 and 6**- The introduction of aromatic substituents into positions 2, 4 and 6, particularly halogenated (i.e., chloride, β-ethyl, etc.) which can increase biological activity[17].

Reactivity

The chemical properties of imidazole contain aspects that are similar to both pyridine and pyrrole. The nitrogen at N-3 exhibits a free electron pair that is not part of the aromatic system; this nitrogen will readily react when exposed to electrophilic attack. Conversely, the nitrogen that is analogous to pyrrole is incorporated into the aromatic sextet so does not undergo electrophilic reaction.

While the imidazole ring has the ability to endure electrophilic substitutions at the carbon locations within the imidazole ring, it is fairly resistant to undergoing nucleophilic substitutions barring instances where there are strongly electron withdrawing substituents already on the imidazole ring. The site that is most likely to undergo nucleophilic attack on an unactivated imidazole ring is the C-2 position; however, when benzimidazoles are formed, the attached aromatic ring (benzene) withdraws from the C-2 position and increases the reactivity of this position with respect to nucleophiles. The increase in reactivity at the C-2 position of benzimidazoles, compared to imidazoles, allows for a greater variety of nucleophilic substitution reactions, compared to imidazole.

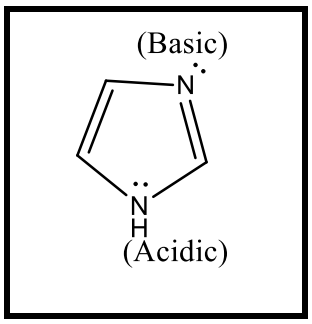


Fig:14

Imidazole and benzimidazole can be analyzed based on their different resonant structures (i.e. dipolar forms have an equally important contribution). The properties of both imidazole and benzimidazole lead to the following conclusions:[18]

- Electrophilic substitution can take place either on the N-3 or any of the carbon atoms in the imidazole ring.

- Nucleophilic attack is most likely at C-2 (or possibly at C-1 in imidazole).
- The molecules exhibit amphoteric behaviour, which means they may function as both acids and bases.

Nucleophilic activity at C-2 of benzimidazole is enhanced by the resonance contributors' pdf as seen in the ionized state, with the electronegative nature of the benzene leaving the nucleophile to react more strongly to C-2 than would be expected if there was no benzene present.

Biological Properties of Imidazole

Imidazoles are well-known heterocyclic compounds that are often found and play a significant role in a number of therapeutic medicines. Several literature reviews indicate that imidazole derivatives have a range of pharmacological activities:

1.Antifungal Activity

2.Anti Tubercular Activity

3.Anti Bacterial Activity

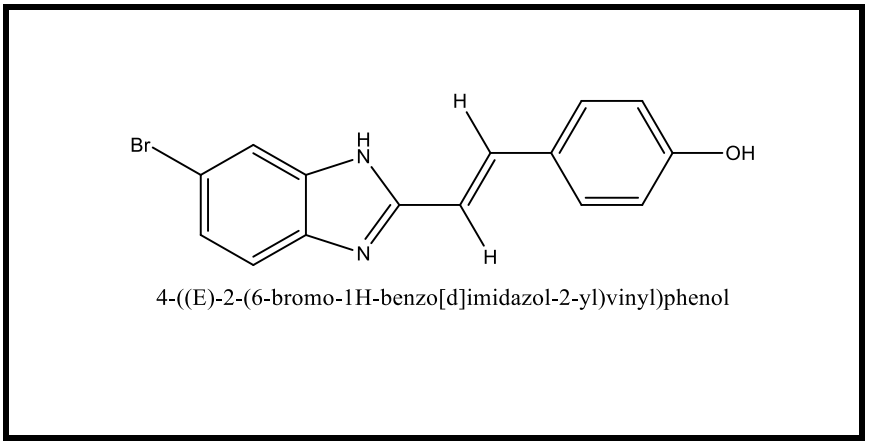
4.Anti Cancer Activity

5.Anti Inflammatory and Analgesic Activity

6. Anti-HIV Activity

1. Antifungal and antibacterial activity:

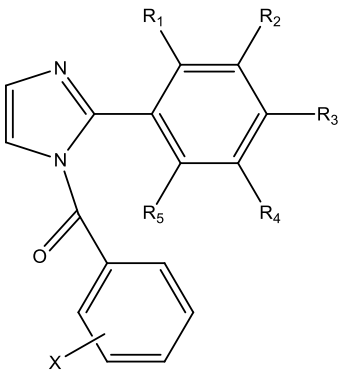
Ramya V *et al.* (2010) synthesized a variety of new 5-(nitro/bromo)-styryl-2-benzimidazole derivatives and tested their antibacterial and antifungal activities against Staphylococcus aureus, Escherichia coli, Enterococcus faecalis, and Klebsiella pneumoniae, which were comparable to ciprofloxacin.[19]



4-((E)-2-(6-bromo-1H-benzo[d]imidazol-2-yl)vinyl)phenol

Fig:15

Deepika Sharma *et al.* (2010) produced 2-(substituted phenyl)-1H-imidazole and (substituted phenyl)-[2-(substituted phenyl)-imidazol-1-yl]-menthanone analogs and tested their antibacterial activity against Gram-positive, Gram-negative bacteria, and fungal species. Norfloxacin was used as the reference standard, and one molecule emerged as the most powerful.[20]

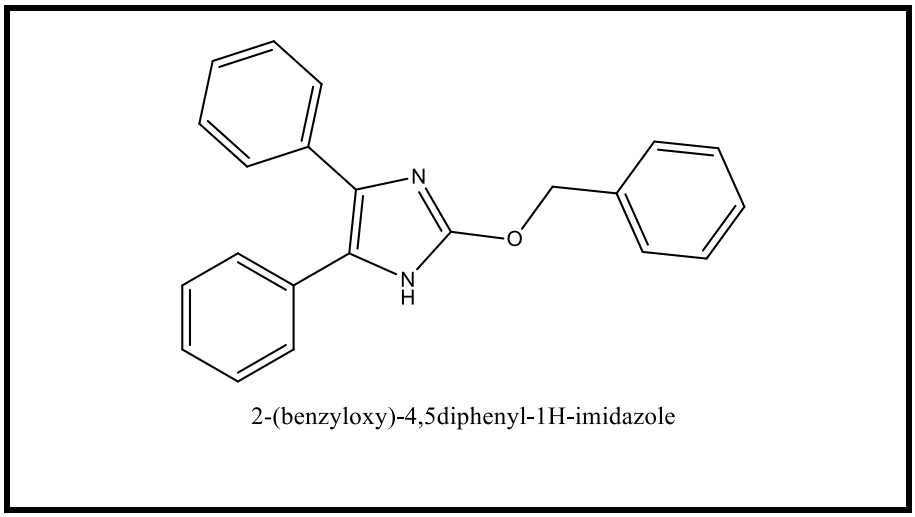


For Compound 1. R1 = Cl, R2 = H, R3= H, R4=H, R5= H,X=4-NO2
2. R1= COOH, R2=H, R3=H,R4=H,R5=H,X=4-NO2
3. R1=H, R2=H, R3=Cl,R4=H,R5=H,X=2-Br
4. R1=H,R2=H,R3=NO2,R4=H,R5=H,X=2-Br

Fig:16

2. Anti-inflammatory and analgesic properties

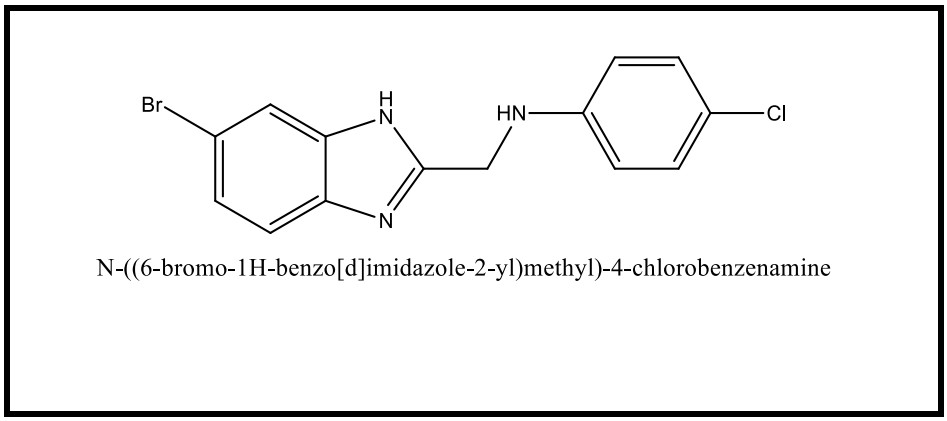
In 2010, Puratchikody and colleagues used the carrageenan-induced paw edema model to investigate the anti-inflammatory properties of 2-substituted-4,5-diphenyl-1H-imidazoles. One molecule had the maximum activity, with indomethacin acting as a reference standard.[21]



2-(benzyloxy)-4,5diphenyl-1H-imidazole

Fig:17

Kavitha C.S. *et al.* (2010) developed a number of 2-methylaminibenzimidazole derivatives, which were then investigated for analgesic and anti-inflammatory characteristics. When compared to standard nimesulide medicine, this compound possesses analgesic properties.[22]

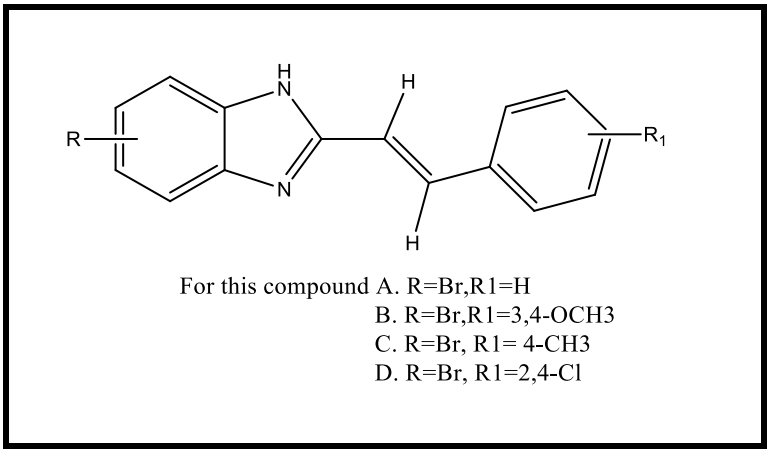


N-((6-bromo-1H-benzo[d]imidazole-2-yl)methyl)-4-chlorobenzenamine

Fig:18

3. Antitubercular activity:

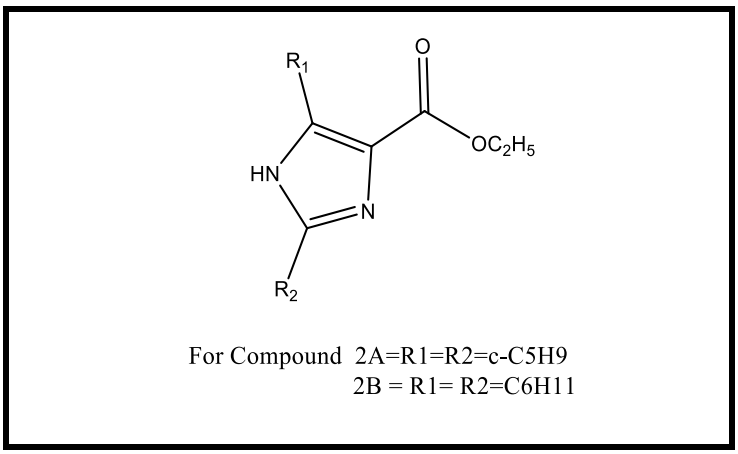
In 2010, Ramya V. and colleagues investigated the in vitro anti-tubercular activity of new 5-(nitro/bromo)-styryl-2-benzimidazole derivatives (1-12) against Mycobacterium TB and found that these compounds were effective. Streptomycin was used as the reference medication.[19]



For this compound A. R=Br,R1=H
B. R=Br,R1=3,4-OCH3
C. R=Br, R1= 4-CH3
D. R=Br, R1=2,4-Cl

Fig:19

In 2010, Preeti Gupta and colleagues studied the anti-TB efficacy of ring-substituted 1H-imidazole-4-carboxylic acid derivatives and 3-(2-alkyl-1H-imidazole-4-yl) propionic acid derivatives against Mycobacterium tuberculosis strains that were both medication-sensitive and treatment resistant. Compounds 2A and 2B emerged as the most powerful.[23]



For Compound 2A=R1=R2=c-C5H9
2B = R1= R2=C6H11

Fig:20

4. Anti-Cancer Activity:

Yusuf Ozkay *et al.* (2010) studied the anticancer properties of newly synthesized imidazole-(benz)azole and imidazole-piperazine compounds. The screening findings revealed that they were the most powerful in the series. Cisplatin served as the reference standard.[24]

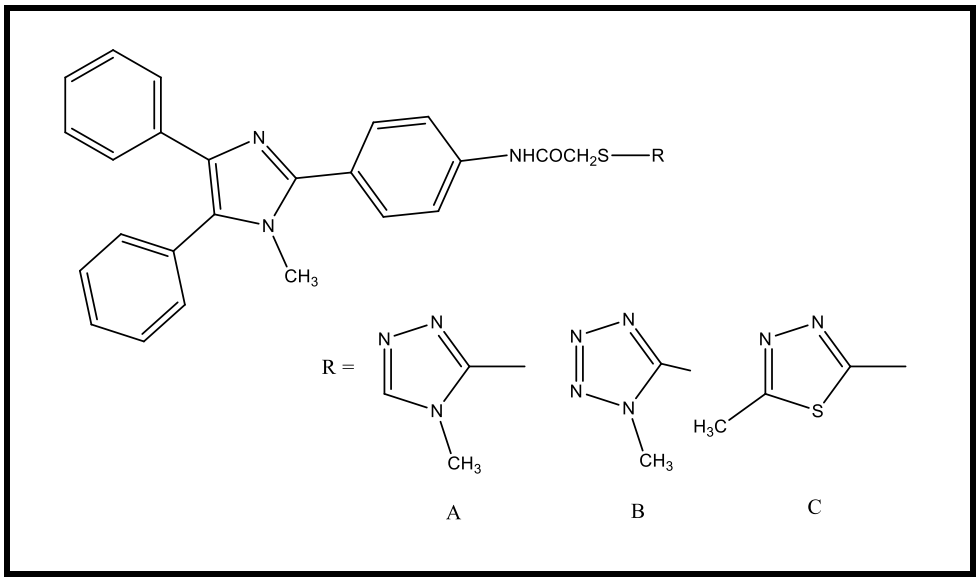


Fig:21

Cenzo Cong iu *et al.* (2008) synthesized and tested several 1,4-diarylimidazole-2(3H)-one derivatives and their 2-thione analogs for anticancer activity. These chemicals showed interesting anticancer effects.[25]

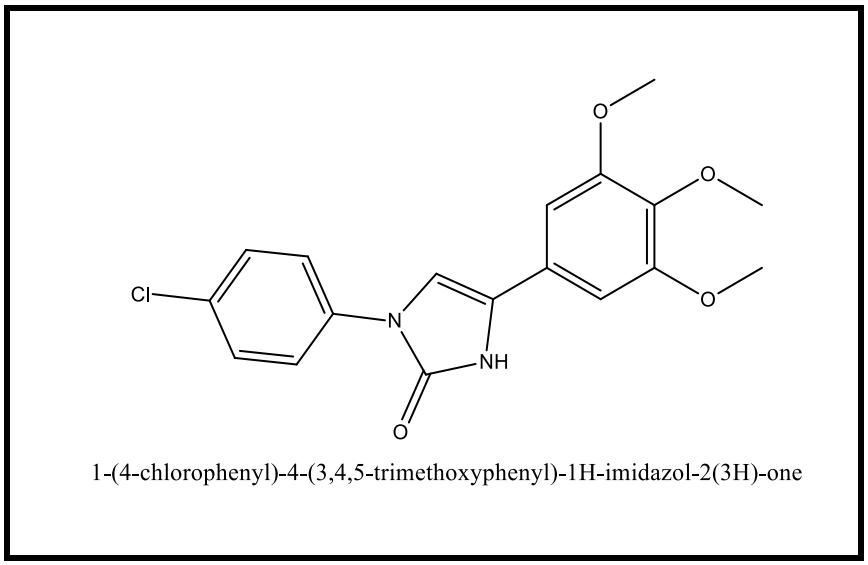


Fig:22

Imidazole's biological significance

Imidazole is a component of several biological substances. The most essential amino acid is histidine, which has an imidazole side chain. Histidine is found in a variety of proteins and enzymes and is required for the structure and binding characteristics of haemoglobin. Histamine is a typical biological compound formed when histidine is decarboxylated. It is

part of the toxin that causes the allergic reaction known as urticaria. The diagram below shows the link between histidine and histamine.

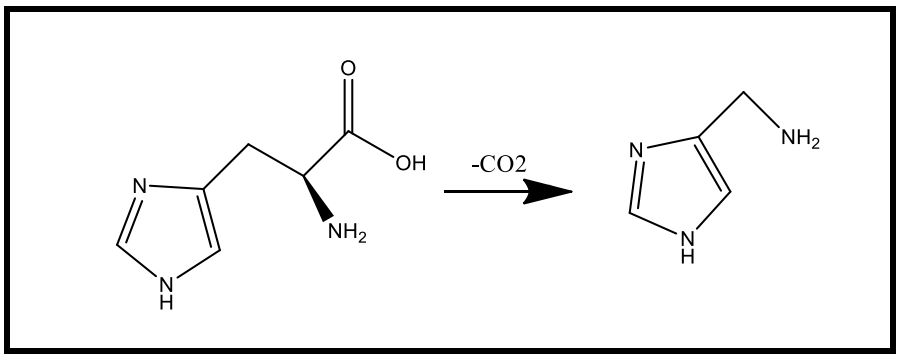


Fig:23

Imidazole is a five-membered aromatic heterocyclic molecule with two nitrogen atoms at positions one and three. Because of its amphoteric character, aromatic stability, and capacity to engage in hydrogen bonding and metal coordination, it is essential in many biological systems and pharmaceuticals.

Applications

Include the purification of His-tagged proteins via immobilized metal affinity chromatography (IMAC). To elute proteins with tags connected to Ni ions attached to the Imidazole is used on the surface of the beads in the chromatography column. His-tagged proteins are released from nickel coordination when an excess of imidazole moves through the column.

Oral imidazole is effective for psoriasis and seborrheic dermatitis. Psoriasis improves in 1.5 to 3 months while seborrheic dermatitis improves in 4-6 weeks, with patients experiencing decreased redness and itching and flaking.The benefits of this medication can be earned without using ointments or creams or using any topical medications.

Imidazole is able to be used at room temperature as a buffer to create solutions with pH 6.2-7.8. It is a buffer for positive hornet peroxides in water. It is also used as a binding agent (chelator) for the divalent metallic ions.[26]

Imidazole is also used in the industrial setting on transition metals such as copper to inhibit corrosion. Corroded copper will lose some of its ability to conduct electricity. Numerous industrial and technical chemicals contain imidazole derivatives.The fire retardant is a polynitroso polybenzimidazole (PBNI) that has an imidazole moiety attached directly to a benzene ring. Imidazole is also used in a variety of electrical and photographic chemicals.

The imidazole nucleus is an important strategy in drug design. A number of pharmaceutical agents have been developed that contain imidazole moieties (e.g., Azomycin, Clotrimazole, Miconazole, Ergothioneine, Clonidine, and Moxonidine). Derivatives of imidazole are among the most significant applications of this compound, having been studied for use as an agent for the treatment of denture stomatitis [27,28].

Imidazole has been incorporated into many types of drugs. It has been used to develop numerous antifungal, antiprotozoal, antihypertensive, and fungicidal drugs that contain synthetic imidazoles. Theophylline contains imidazole, activates the central nervous system, and is frequently found in both tea and coffee. Imidazole is also found in mercaptopurine, which is used to treat leukemia by inhibiting DNA function.

CONCLUSION

Imidazole is an exceptionally important nitrogen-containing heterocyclic compound that is found in many medicines. Because it has a unique aromatic structure that possesses both an acidic and basic character (amphoteric), along with the capacity to form strong hydrogen bonds, imidazole has proven to be an ideal pharmacophore for developing drugs. Many derivatives of imidazoles exhibit a variety of biological activities including antibacterial, antifungal, anticancer, anti-inflammatory, antitubercular**,** antiviral and antiulcer activity. The most common synthetic routes to prepare imidazole derivatives include Debus-Radziszewski synthesis, Van Leusen synthesis, and Phillips method. Numerous clinically important drugs including metronidazole, clotrimazole, ketoconazole and omeprazole contain an imidazole nucleus indicating the therapeutic significance of imidazole. Because of its many pharmacological uses and the ongoing development of new drugs, imidazole is one of the most important heterocyclic scaffolds in the modern pharmaceutical sciences.

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The authors declare no competing financial interests, affiliations or personal relationships that could have appeared to influence the work, findings or conclusions reported in this paper.

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