1.1 Introduction to heterocycles

Heterocycles are cyclic organic compound containing at least one heteroatom such as N, O, S, P, Mg, Fe and Se in the parent scaffold (Figure **1.1**). In general, it is the heteroatom that imparts striking and distinctive properties to heterocycles [1]. It is interesting to note that nearly 50% of known organic compounds and 90% of active pharmaceuticals contain heterocyclic moieties in their structures [2]. This emphasizes the significance of heterocycles in modern drug design. Recent survey has witnessed encouraging growth in the study of heterocyclic compounds due to their numerous pharmaceutical and industrial applications [3]. Many biological compounds such as vitamins, hormones, proteins, amino acids which are associated with living organisms composed of heterocyclic scaffolds. For example, the photosynthesizing pigment; chlorophyll, oxygen transporting pigment; haemoglobin, essential amino acid; proline, vitamins like riboflavin all represent heterocyclic moieties in their structure. They serve as an integral part of a broad variety of biologically active natural products and synthetic drugs [4,5]. Heterocycles can be classified into aromatic and non-aromatic structures. Aromatic heterocyclic compounds containing cyclic aromatic rings represent structural motifs found in a large number of biologically active synthetic and natural compounds [6].

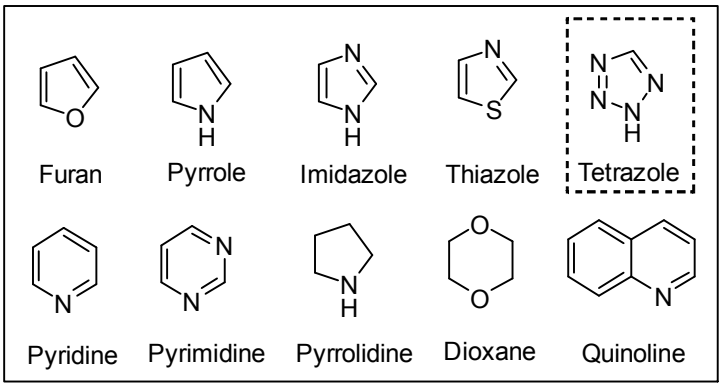

Figure **1.1** Common heterocycles found in drug molecules

Additionally, aromatic heterocycles play a significant role in the synthesis of dyes and polymeric materials of commercial importance. Among different kinds of heterocyclic ring systems, nitrogen containing heterocyclic motifs are most abundant due to its wider presence in a large number of bioactive molecules [7].

They also finds widespread applications among fungicides, herbicides and other application-oriented molecules [2]. In particular, important ring systems like furan, imidazole, pyrimidine, quinoline, tetrazole and their benzo-fused analogues are associated with a wide range of biological activities [1]. The ability of the nitrogen atom to form intermolecular forces such as hydrogen bonding, dipole-dipole interactions, hydrophobic effects and van der Waals forces have enhanced their utility in various therapeutic applications [8].

In our quest and endeavour to provide more in-depth insight into nitrogen containing aromatic heterocycles; we have divided this discussion into five sections: (i) three- and four-membered rings; (ii) five-membered rings; (iii) six-membered rings; (iv) seven- and eight-membered rings; and (v) fused rings. The relative presence of various nitrogen heterocycles vary significantly such that six-membered rings (59%) are the most frequently utilized, followed by five-membered (39%) and fused rings (14%) [2].

1.1.1 Four membered aromatic nitrogen heterocycles

The nitrogen heterocycles in this sub-class are mostly β-lactams, of which 95% are fused to another ring through the shared nitrogen atom. For example, Ezetimibe of the β-lactam family is a medicine used for the treatment of high blood cholesterol and other lipid abnormalities (Figure **1.2**) [9,10].

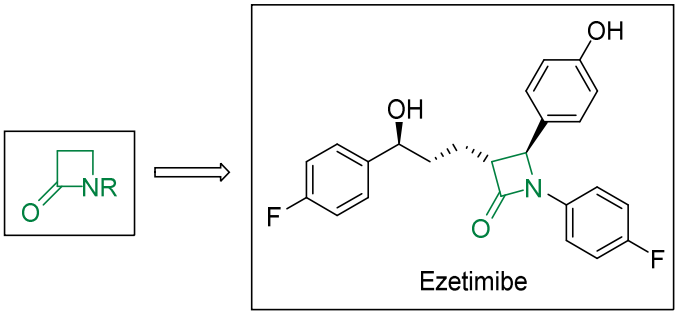

Figure **1.2** Drug molecules containing four-membered aromatic nitrogen heterocycles

1.1.2 Five membered aromatic nitrogen heterocycles

These aromatic heterocycles appear in a total of 101 drugs, which is approximately 9% of the total number of Food and Drug Administration (FDA) approved small

molecules. In this five-membered aromatic nitrogen heterocycles, only indole molecule contains a single nitrogen atom while others have additional nitrogen (imidazole, tetrazole, benzimidazole, and thiazole) (Figure **1.3**). Nizatidine (drug for peptic ulcer), Etomidate (intravenous anaesthetic agent), Emedastine (antihistamine used in eye drops), Pemirolast (antiallergic); all contain five membered nitrogen heterocycles in their structure (Figure **1.3**) [11,12].

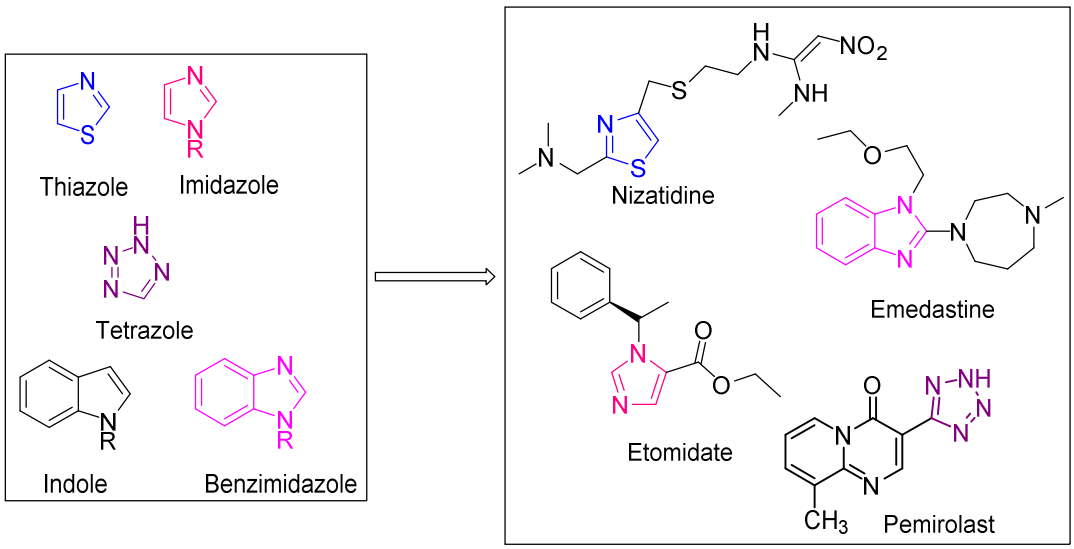

Figure **1.3** Drug molecules containing five-membered aromatic nitrogen heterocycles

1.1.3 Six membered aromatic nitrogen heterocycles

In this category, heterocycles contain a single or additional heteroatom, which in almost all cases is a nitrogen atom (pyrimidine, quinazoline, and pyrazine).

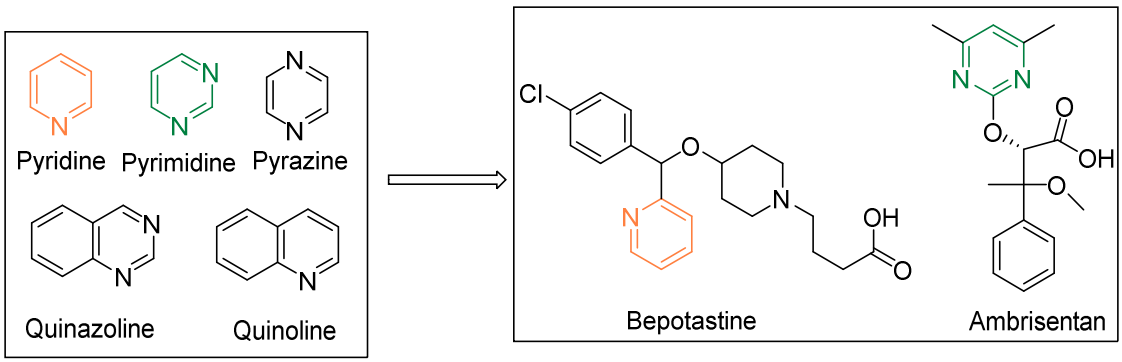

Figure **1.4** Drug molecules containing six-membered aromatic nitrogen heterocycles

Pyridine and quinoline contain only one nitrogen atom whereas pyrimidine, pyrazine, quinazoline constitute more than one nitrogen atom. Bepotastine (antihistamine) and Ambrisentan (antihypertensive) are two drug molecules having pyridine and pyrimidine moieties in their structures respectively (Figure **1.4**) [13,14].

1.1.4 Seven and eight-membered aromatic nitrogen heterocycles

Although less common compared to their five and six-membered ring counterparts, seven and eight-membered nitrogen heterocycles are important pharmaceutical core fragments. Most commonly found heterocycles in this category are shown in (Figure **1.5**). Oxcarbazepine, under the brand name 'Trileptal' is used to treat epilepsy [15].

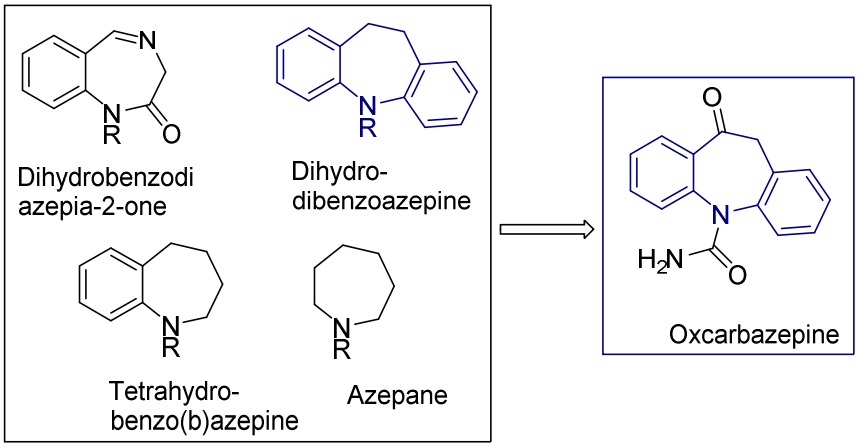

Figure **1.5** Drug molecules containing seven and eight-membered nitrogen heterocycles

1.1.5 Fused nitrogen heterocycles

Fused ring systems which we define as those nitrogen heterocycles that contain more than one nitrogen heterocycle, although not necessarily fused directly adjacent to each other. An antiviral drug, Famciclovir contains fused nitrogen heterocycles in their structure (Figure **1.6**) [16].

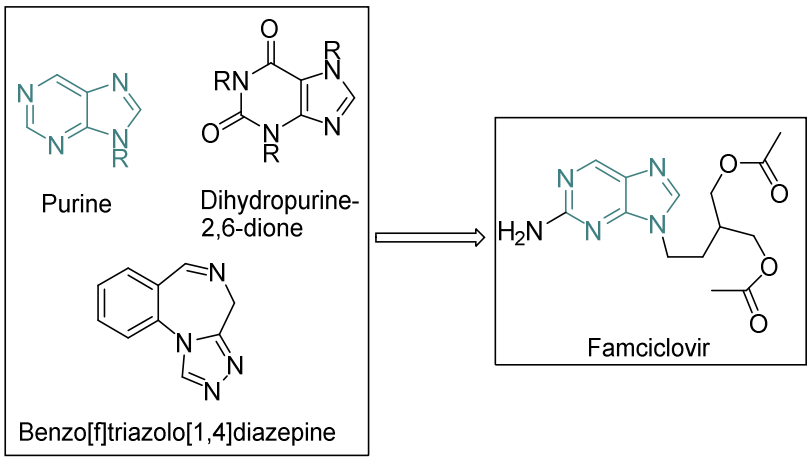

Figure **1.6** Pharmaceuticals containing fused aromatic nitrogen heterocycles

1.2 The importance of *N*-heterocycles in biochemistry and life processes

Heterocyclic compounds accounts for nearly half of the known organic compounds. Among them, nitrogen containing heterocycles are of great importance to life science; since they are abundant in nature, existing as subunits in several natural products [5]. Vitamins, nucleic acid, enzymes, co-enzymes, hormones and alkaloids all contain *N*-based heterocycles in their structure. The prevalence of *N*-heterocycles in biologically active compounds can be attributed to their stability and operational efficiency in human body. Heterocycles play a critical part in biochemical processes [17]. The most important and essential constituents of living cells; DNA and RNA, which carry the genetic information are made up of pyrimidine (cytosine, uracil and thymine) and purine (adenine and guanine) bases which consist of aromatic nitrogen heterocycles (Figure **1.7**) [18].

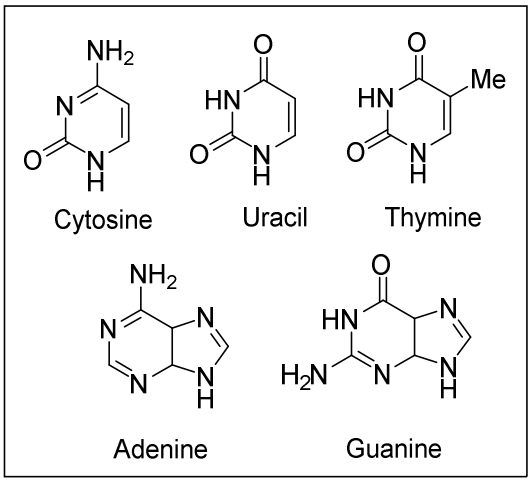

Figure **1.7** Pyrimidine and Purine bases

The essential amino acids tryptophan and histidine are made up of aromatic nitrogen heterocycles. They participate with other amino acids in protein constitution through amide linkages. Histamine, formed by decarboxylation of histidine is a powerful vasodilator released in allergic responses while serotonin formed from tryptophan, is an important neurotransmitter (Figure **1.8**) [4].

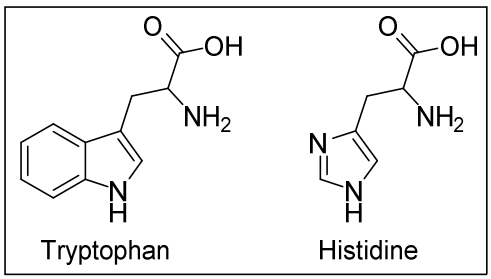

Figure **1.8** Heteroaromatic amino acids

Enzymes possess protein structures entirely; coenzymes incorporate non-amino acids in their structures [19]. Most coenzymes have aromatic nitrogen heterocycles as major constituents and are essential for the redox biochemical transformations. For example, nicotinamide adenine dinucleotide (NAD) and flavin adenine dinucleotide (FAD) (Figure **1.9**) [20].

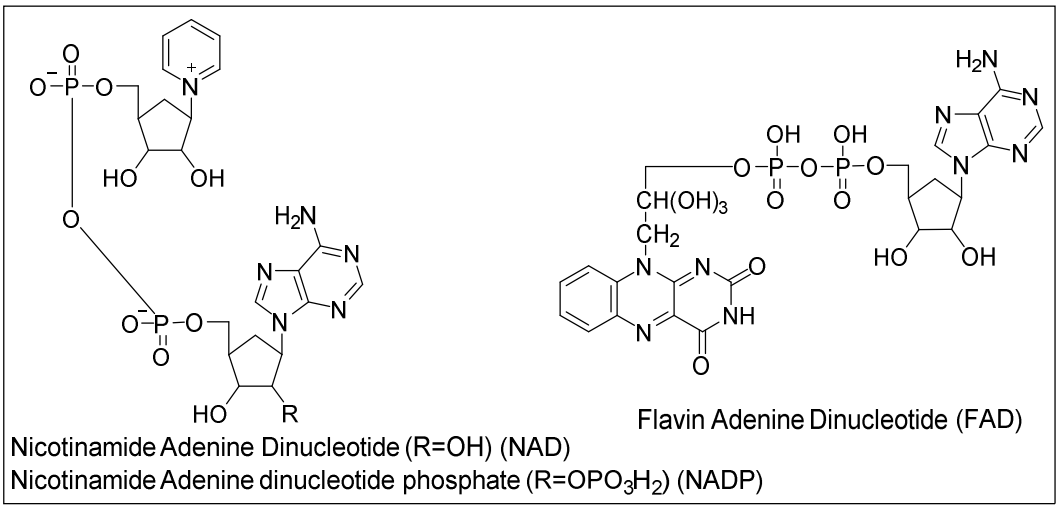

Figure **1.9** Coenzymes

Pyrroles are five-membered nitrogen-containing heterocyclic compounds present in heme, a constituent of haemoglobin. Haemoglobin is an iron containing protein which transports oxygen in our body [21]. Each haemoglobin molecule is made up

of four heme groups surrounding a globin group and forms a tetrahedral structure (Figure **1.10**) [2].

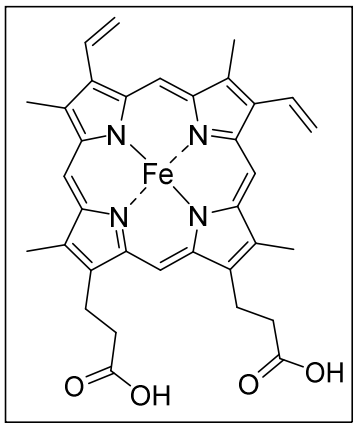

Figure **1.10** Structure of Haemoglobin

Vitamins are organic compounds that our body use, in very small amounts, for a variety of metabolic processes. Indispensable diet components such as vitamin E, vitamin C, nicotinamide (vitamin B_3); containing *N*- and *O*- based heterocycles (Figure **1.11**) [4].

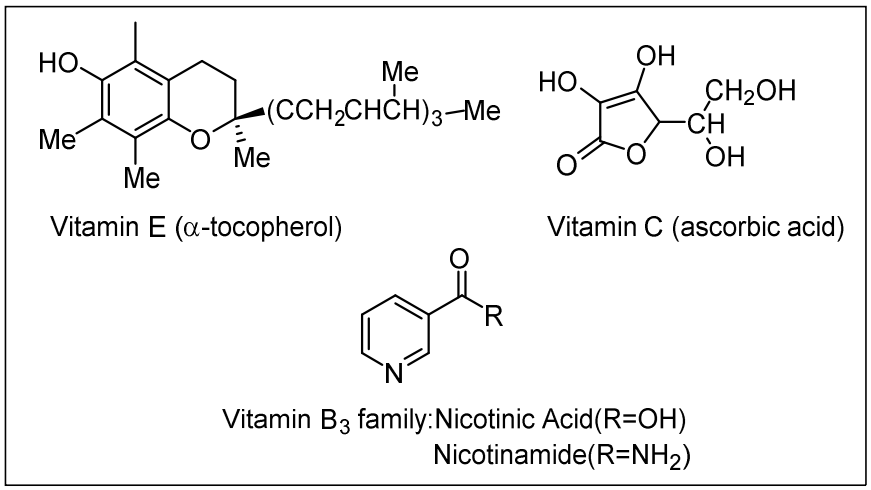

Figure **1.11** Vitamins containing heterocyclic moieties

1.3 Applications of nitrogen-containing heterocycles

Heterocyclic compounds make up a large portion of FDA approved medicines and have been proven to possess anti-diabetic, antibacterial, anticancer, antiviral, antimicrobial, anti-inflammatory, anti-hypertension, anti-malarial, anti-Alzheimer's and antifungal activities [22]. Analysis of database reveals that about 60% of the

FDA approved pharmaceuticals are nitrogen-based heterocycles. They form an integral part in many biologically active molecules including dyes, agrochemicals amongst others.

1.3.1 Anticancer activity

Heterocyclic structures play a key role featuring prominently in anticancer drugs currently available in the market. An anticancer medication is a therapy effective in treating malignant or cancerous disease. Anastrozole and Gemcitabine are anticancer drugs available in the market for the treatment of breast and lung cancer (Figure **1.12**). Anastrozole contain a triazole functional group and act as competitive inhibitors perturbing the catalytic properties of the hemeprosthetic group [23,24].

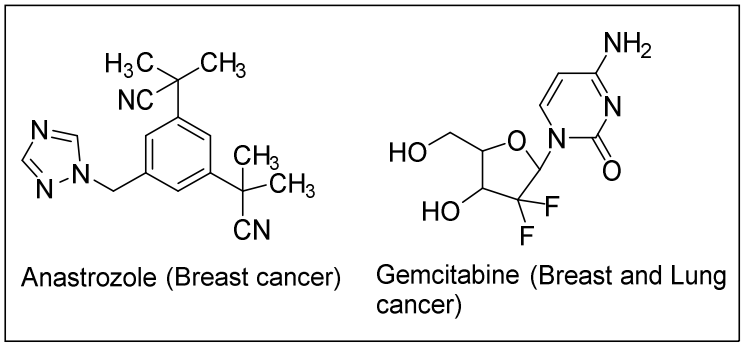

Figure **1.12** Anticancer drugs in clinical use

1.3.2 Anti-inflammatory activity

The property of a medicine that lowers or reduces inflammation or swelling is referred to as an anti-inflammatory agent. These chemicals are known as "non-steroidal anti-inflammatory drugs" (NSAIDs). Etoricoxib and Indometacin are effective anti-inflammatory drugs (Figure **1.13**) [25]. The importance of the 2′-methyl group in indole of Indometacin prompted to explore other functionality at this position. For instance, 2′-methyl group when replaced by a H-atom resulted in a decrease in its anti-inflammatory activity [26,27].

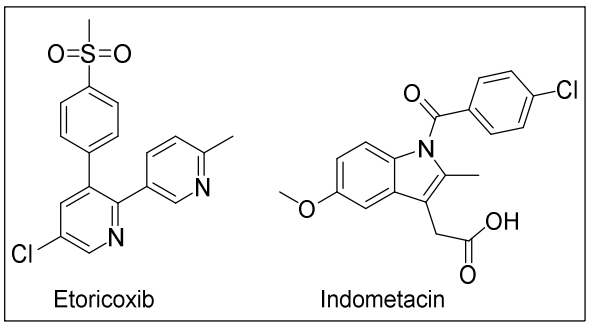

Figure **1.13** Anti-inflammatory drugs in clinical use

1.3.3 Antibacterial activity

Antibacterial or antibiotics is a term used to describe drugs used for prevention or treatment of bacterial infections, either by killing or inhibiting the growth of bacteria. Amoxicillin and Ciproflaxin are common antibacterial drugs (Figure **1.14**) [28,29].

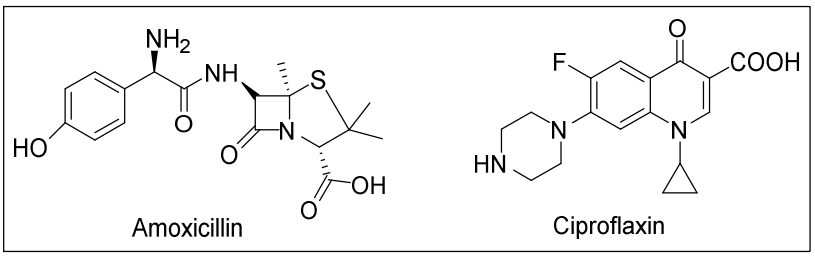

Figure **1.14** Antibacterial drugs in clinical use

1.3.4 Antiviral activity

Viruses have the simplest biological structure, comprising of various RNA or DNA strings and an outer protein layer that can be enveloped by a lipid coating. Viruses can cause an immunological response in the human host, which can regulate the infection resulting in pathological symptoms and even death. Antiviral drugs are used to treat viral infections. For example, Zanamivir and Xofluza are antiviral drugs (Figure **1.15**) [30-32].

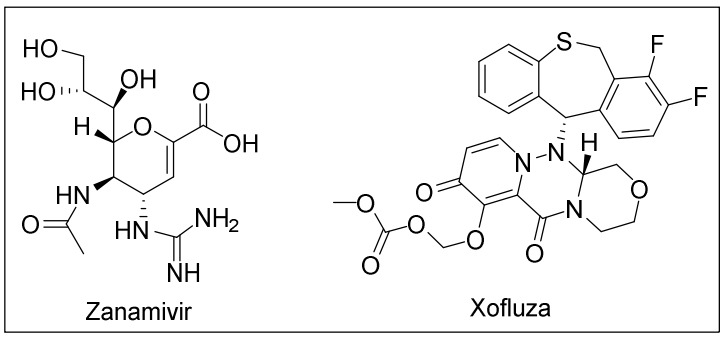

Figure **1.15** Antiviral drugs in clinical use

1.3.5 Anticonvulsant activity

Anticonvulsants are drugs or pharmacological agents that are used to control seizures or prevent sequence of seizures from happening. These are defined as the agents that prevent the severity of convulsive seizures. For instance, Perampanel and Clobazam are anticonvulsant drugs (Figure **1.16**) [33,34].

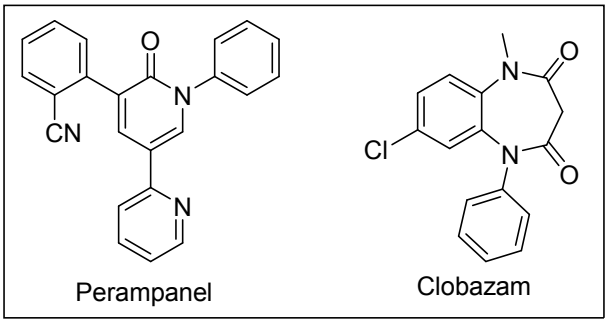

Figure **1.16** Anticonvulsant drugs in clinical use

1.3.6 Anti-leprosy activity

Various skin infections, including dermatitis herpetiformis, and leprosy; both are treated using anti-leprosy drugs. Clofazimine and Ethionamide are anti-leprosy drugs (Figure **1.17**). The key structural feature of Clofazimine which has an isopropylimino group at position 2 of the phenazine nucleus reveals that alkylimino group is critical for antimicrobial activity [35,36].

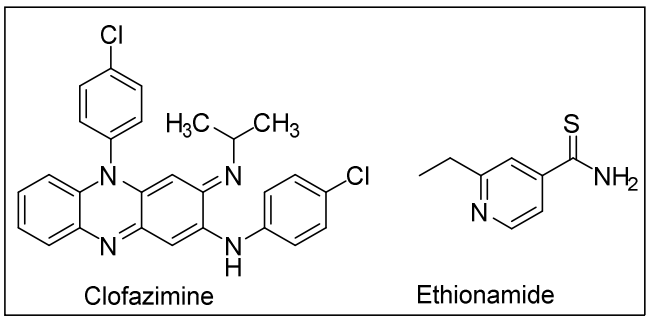

Figure **1.17** Anti-leprosy drugs in clinical use

1.3.7 Antifungal activity

Substances that are used to treat fungal infection which is most commonly located on the skin, hair and nails are called antifungal drugs. Fluconazole and Clotrimazole are triazole and imidazole containing antifungal drugs respectively (Figure **1.18**) [37,38].

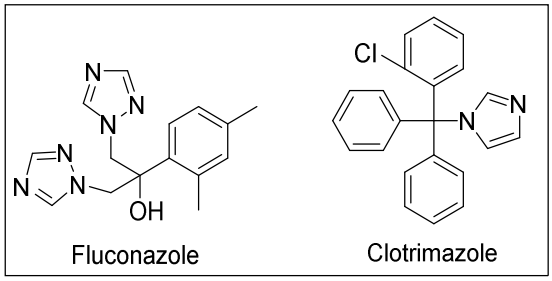

Figure **1.18** Antifungal drugs in clinical use

1.3.8 *N*-heterocycles as agrochemicals

Nitrogen containing heterocycles have arisen as one of the prominent agents for crop protection. Pinoxaden is a new herbicide used to reduce the growth of grass weeds in cereals.

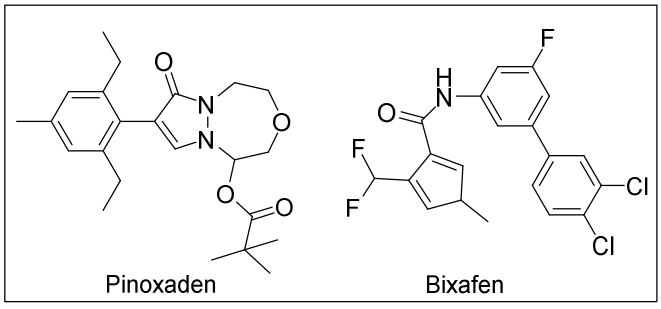

Figure **1.19** Nitrogen heterocycle based agrochemicals

Similarly, another nitrogen-containing fungicide, Bixafen shows significant activity against a broad spectrum of cereal diseases (Figure **1.19**) [39,40].

1.3.9 *N*-heterocycles as dyes

Nitrogen-bearing heterocyclic dyes have varied applications in diverse fields like analytical chemistry, pharmaceuticals, textiles, cosmetics and leather industry. Considering the relevance of colorants in modern times, dyes have acquired a distinct position in synthetic organic chemistry. For example, the important vat dye, indigo which contains two indole units, is widely used for colouring cellulosic fibre, especially cotton fibre along with viscose rayon, leather and other fibres (Figure **1.20**) [41].

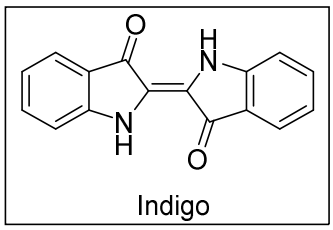

Figure **1.20** Dyes containing nitrogen heterocycles

1.3.10 *N*-heterocycles as catalysts

Various transition metal complexes with *N*-heterocyclic carbene (NHC) ligands have been used in catalytic research and cover a broad domain of reactions ranging from olefinic polymerization to C-C coupling. NHCs are neutral, having high nucleophilicity, two-electron-giving ligands with a heterocyclic moiety mainly comprising of pyrazole, triazole, tetrazole, imidazole and benzimidazole. Ruthenium-catalysed (Grubb's catalyst 2.0) olefin metathesis is one of the most important and well-studied reactions mediated by NHC–metal complexes (Figure **1.21**) [42].

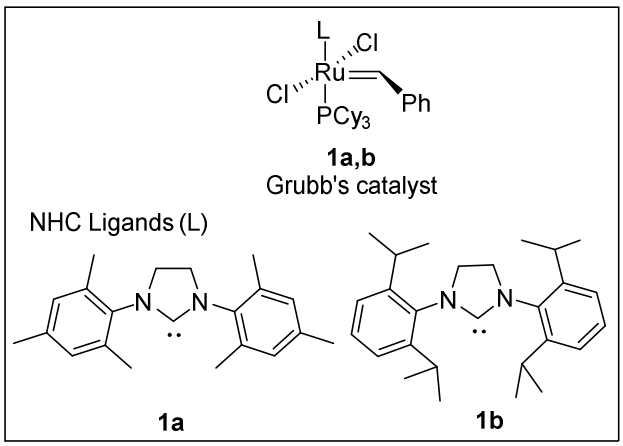

Figure **1.21** Catalyst containing nitrogen heterocycles

1.3.11 *N*-heterocycles as corrosion inhibitors

Iron and its alloys are essential and are being extensively used in industries. Corrosion inhibitors ensure that there is no or less metal dissolution. *N*-heterocycles like 4-chloro-1*H*-pyrazolo[3,4-*d*]pyrimidine in 1M HCl exhibited good anti-corrosion activity for mild steel. Benzothiazole showed corrosion inhibition efficiency on stainless steel in 3M H_2SO_4 (Figure **1.22**) [43].

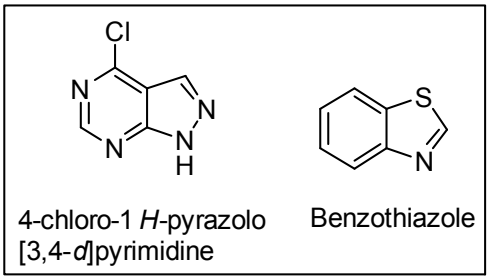

Figure **1.22** Corrosion inhibitors containing nitrogen heterocycles

1.4 Conclusion

Nitrogen-containing heterocyclic compounds are one of the important sorts of organic compounds, which are taking a wide range of applications in various fields. Today there are a lot of heterocyclic compounds known showing applications in medicinal chemistry, cosmetics industry, food industry, biochemistry. Heterocyclic compounds have a role in most fields of sciences and number is increasing rapidly due to the enormous synthetic utility.

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