This chapter mainly gives the information about the conclusions of the present work. In the present work, we have synthesized the different $N$-containing heterocyclic compounds like BDPs, DHPMs and 3,4-dihydroquinoxalin-2-amine derivatives using simple, inexpensive, eco-friendly, commercially scalable catalysts and studied the biological activity for all compounds. Some of the compounds showed the significant anti-neuroinflammatory activity. The utility of AlKIT-5 catalyst for synthesis of BDPs, DHPMs, 3,4-dihydroquinoxalin-2-amine derivatives would be precious addition to available methods. We have demonstrated for the first time the use of 3D mesoporous aluminosilicate catalyst with cage type pore for the synthesis of BDPs, DHPMs and 3,4-dihydroquinoxalin-2-amine derivatives. This method is quite simple and heterogenous. The catalyst gave high isolated yield of the derivatives of BDPs and DHPMs in a shorter reaction time and can be recycled several times. We strongly hope that the highly stable AlKIT-5 catalyst could pave the way for the production of BDPs, DHPMs and 3,4-dihydroquinoxalin-2-amine derivatives and create the platform for the commercialization of the process by replacing the existing homogenous catalysts which suffered from various drawbacks such as corrosion, toxicity, waste production and a high cost. We found AlKIT-5(10) is superior over the other catalysts such as silica gel-supported sulfuric acid, DDQ and PTSA.
**AlKIT-5 catalyst:** A Novel three dimensional cubic $Fm3m$ mesoporous aluminosilicates (AlKIT-5) with very high structural order and unprecedented loadings of Al in the silica framework have been successfully prepared and used in the synthesis of various biologically active compounds such as BDPs and DHPMs and 3,4-Dihydro-quinoxalin-2-amine derivatives. The acidity and the catalytic activity of the AlKIT-5 catalysts have been studied and the results were compared with the other catalysts. It has been found that the AlKIT-5 catalysts are highly active and the AlKIT-5(10) is superior over the other catalysts such as AlKIT-5 catalysts with the $n_{Si}/n_{Al}$ ratio 28, 44. AlKIT-5(10) is also more active than the reported catalysts such as mordenite, zeolite H-Y, zeolite H-b, and ZSM-5. Among the catalysts examined, AlKIT-5(10) is found to be superior, scalable over the other catalysts such as silica gel supported sulfuric acid, PTSA and DDQ reagent.

**BDPs:** We have demonstrated for the first time the synthesis of 1,5-benzodiazepines using silica gel supported sulfuric acid and AlKIT-5 catalysts through a condensation reaction between substituted OPDA and a series of symmetrical and unsymmetrical ketones at room temperature in acetonitrile solvents. The two catalysts were found to be highly active. But AlKIT-5 showed better performance and affording a high yield of benzodiazepines. The effect of the aluminium content of the catalyst and the catalyst concentration on the above process was investigated. The catalyst was also successfully employed for the
preparation of various derivatives of 1,5-benzodiazepine using substituted OPDAs and various ketones. In all cases, the reactions are highly selective and are completed within 1.0–2.5 h. The catalyst showed excellent activity in all the cases, showing 85–97% isolated yield of the corresponding derivatives of 1,5-benzodiazepine. The high activity of the catalyst is mainly due to its high acidity, excellent textural parameters such as high surface area, large pore volume, and cage type 3D porous structure. This method is quite simple and selective. The catalyst gave high isolated yield of the derivatives of 1,5-benzodiazepine in a shorter reaction time at room temperature and can be recycled several times. We strongly hope that the highly stable AlKIT-5 catalyst could pave the way for the production of 1,5-benzodiazepine and its derivatives and create the platform for the commercialization of the process by replacing the existing homogenous catalysts which suffered from various drawbacks such as corrosion, toxicity, waste production, and a high cost.

**BDPs Merits:**

1. Synthesis of benzodiazepines derivatives have attracted prominent role in the field of drugs and pharmaceuticals.

2. Biologically active chloro substituted benzodiazepines are synthesized.

3. We have used substituted ketones like 2-acetyl and 3-acetyl thiophenes to get Multifunctionalised compounds, which are having high biological activity.
4. Thio and chloro derivatives are possessing high biological activity.

5. AlKIT-5 catalyst is heterogeneous and good acidic catalyst. Simple work up, eco-friendly method, catalyst cost is low, preparation is easy, reusability of catalyst. The reactions proceed smoothly and efficiently. Products are formed in excellent yields in short reaction times.

6. The reactions are clean and do not require any additives or acidic promoters.

7. In addition to its simplicity and milder reaction conditions, this method has ability to tolerate a wide variety of substituents.

8. The procedure has wide applications in the synthesis of substituted compounds.

9. The reaction rates and yields are dramatically enhanced by this catalyst. The high activity of the catalyst is mainly due to its high acidity, excellent textural parameters such as high surface area, large pore volume, and cage type 3D porous structure.

10. Scale up studies (1 g batch): In addition, we have also carried out scale up experiments with 1.08 g of OPDA, (Chapter-III, Table 3.3, entry 1) using AlKIT-5 catalyst and we achieved 97% of the yield which is almost nearer to the reported yield in Table 3.3 (entry 1), revealing that the catalyst could easily be commercialized.

11. The anti-neuroinflammatory studies of the products (3a-t) are investigated. Among them compound 3e shown high activity. For these products, the further clinical, formulation and epicacy studies are needed.
**DHPMs:** In conclusion, we have developed a simple, convenient and effective method for the synthesis of DHPMs and their derivatives using substituted aromatic aldehydes, β-ketoester, urea or thiourea at reflux temperature using DDQ and AlKIT-5 catalyst under acetonitrile conditions. Among the two catalysts AlKIT-5 is superior in terms of yields and recoverability. This method is applicable to a wide range of substrates including aromatic, aliphatic, α,β-unsaturated and heterocyclic aldehydes. The catalyst was found to be highly active and selective, affording a high yield of DHPMs with good recyclability. Compared to the classical Biginelli reaction conditions, this new approach consistently has the advantage of excellent yields (80–96%) and short reaction times, 3.0–4.0 h. The effect of the acidity and the concentration of the catalyst on the above process was investigated. We also demonstrate the synthesis of various multifunctional Biginelli compounds using the highly active AlKIT-5 catalysts. The mesoporous AlKIT-5 catalysts are promising heterogeneous catalysts in all circumstances where the aluminosilicate matrix is highly stable and we strongly hope that this catalyst could also be used for other acid catalyzed organic transformation and help to replace the existing toxic, corrosive and expensive homogenous catalysts.

**DHPMs Merits:**

1. Synthesis of DHPMs derivatives have attracted prominent role in the field of drugs and pharmaceuticals.
2. We have synthesized biologically active thio derivatives of substituted DHPMs.

3. Simple work up, eco-friendly method, catalyst cost is low, preparation is easy, reusability of the catalyst. The reactions proceed smoothly and efficiently. Products are formed in excellent yields in short reaction times.

4. The reactions are clean and do not require any additives or acidic promoters.

5. In addition to its simplicity and milder reaction conditions, this method has ability to tolerate a wide variety of substituents.

6. The reaction rates and yields are dramatically enhanced by AlKIT-5 catalyst.

7. Scale up studies (1 g batch): In addition, we have also carried out scale up experiments with 1.06 g (10 mmol) of benzaldehyde, (Chapter-IV, Table 4.3, entry 4a,) using AlKIT-5 catalyst and we achieved 95% of the yield which is almost nearer to the reported yield in Table 4.3 (entry a), revealing that the catalyst could easily be commercialized.

8. The anti-neuroinflammatory studies of the products (4a-4z & 4a1-4d1) are investigated. Among them 4g, 4j and 4v compounds shown high activity. For these products, the further clinical and formulation epicacy studies are needed.

**3,4-Dihydroquinoxalin-2-amine derivatives:** We have synthesized a novel 3,4-dihydroquinoxalin-2-amine derivatives using various aromatic diamines, carbonyl compounds and diverse isocyanides in
the presence of $p$-toluene sulfonic acid and AlKIT-5 catalysts. Among the two catalysts AlKIT-5 is superior in terms of yields and recoverability. This method is applicable to a wide range of substrates and simple work-up, chromatography-free and products were recrystalised by using ethanol to get high purity. The anti-neuroinflammatory studies of the novel products (4a-p) were investigated and the results are presented in Chapter VI. To the best of our knowledge these particular compounds are not reported by any one in the literature using these two catalysts.

**3,4-Dihydroquinoxalin-2-amine derivatives Merits:**

1. The novel 3,4-Dihydroquinoxalin-2-amine derivatives are first time synthesized in excellent yields using chloro OPDA, diverse ketones and various isocyanides in the presence of PTSA and AlKIT-5 catalysts.

2. Scale up studies (1 g batch): In addition, we have also carried out scale up experiments with 1 g of OPDA (Chapter-V, Table 5.2, entry a) using AlKIT-5 catalyst and we achieved 93.5% of the yield which is almost nearer to the reported yield in Table 5.2 (entry a), revealing that the catalyst could easily be commercialized.

3. The anti-neuroinflammatory studies of the novel products (4a-p) are investigated. Among them 4l and 4n compounds showed high activity. For these products, the further clinical and formulation epicacy studies are needed.
4. Some of the compounds are significantly possessing high anti-neuroinflammatory activity. These compounds may lead for new drug discovery.

**Biological activity:** We have evaluated the anti-neuroinflammatory effect in microglia cells for all synthesized $N$-containing heterocyclic compounds such as BDPs, DHPMs and 3,4-dihydroquinoxalin-2-amine derivatives. Among the various compounds tested, some compounds strongly inhibited LPS-induced nitric oxide (NO) production with IC$_{50}$ value in the microglia cells. These compounds significantly inhibited the enzyme activity of inducible NO synthase (iNOS) without changes of iNOS protein expression and NO scavenging activity. This result suggests that some compounds showed the anti-neuroinflammatory effect by suppressing iNOS. Further studies are required to elucidate precise mechanisms underlying the anti-inflammatory activity of active compounds.

In conclusion, our studies during this thesis work have contributed to the existing knowledge of $N$-Containing heterocyclic compounds in the following aspects. In the present research work, we have synthesized the biologically active Multifunctionalised novel molecules and their basic synthesis, suitable catalyst for commercialization of the process and the anti-neuroinflammatory studies are investigated.
**Future scope of the work:** Since different N-containing heterocyclic compounds like BDPs, DHPMs and 3,4-dihydroquinoxalin-2-amine derivatives are possessing good pharmacological properties, we should explore the synthesis of Multifunctionalised compounds and we should investigate the anti-neuroinflammatory studies. We expect the novel Multifunctionalised compounds should be promising drugs in future.

**List of Publications**

1. Dichloro Dicyano Quinone (DDQ) as coupling reagent for high yield synthesis of 3,4-Dihydropyrimidin-2(1H)-ones. **Shobha, D.;** Chari, M.A.; Sadanandam, P.; Makkanti, K. *Journal of Heterocyclic Chemistry, 45, 2008*, 1225-1227.


