The present thesis describes a study towards the development of newer methodologies for the synthesis of a wide spectrum of pyrimidine derivatives. The targeted pyrimidine scaffolds have been known for their immense biological, medicinal as well as synthetic values. A few of the synthesized compounds are found to be novel and characterized by single crystal X-ray analysis along with other spectroscopic techniques (FT-IR, Mass, ^{1}H & ^{13}C NMR) and CHN analysis. Preliminary screening towards the antibacterial property of some selected pyrimidine derivatives has also been performed and covered in the thesis. The thesis has been organized in five major chapters followed by a brief sixth chapter summarizing the overall conclusion and future scopes.

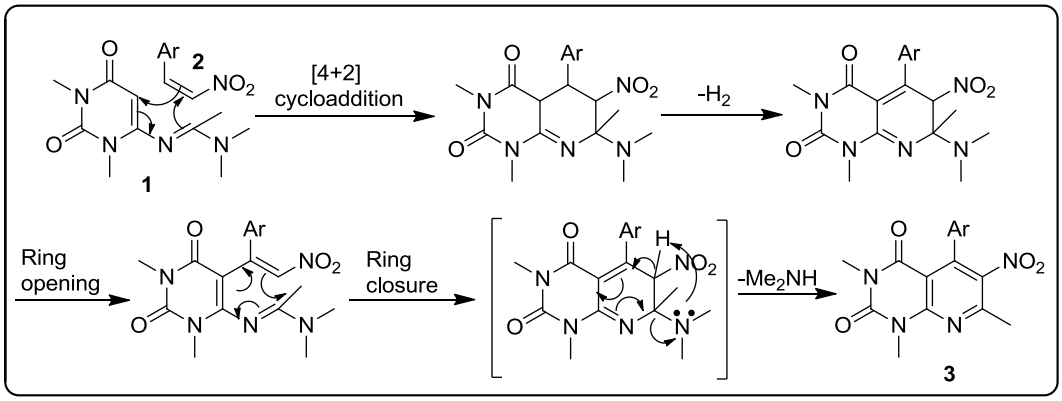
Chapter 1: Introduction & early history

Chapter 1 is a brief introduction to *pyrimidine* chemistry. This chapter is a humble attempt to narrate the history, natural occurrence and importance of *pyrimidines*. Since the discovery, as an important constituent of nucleic acids, *pyrimidines* have been occupying a distinct and unique place in human's life and consequently in organic and medicinal chemistry research. Although, the moiety is mainly known for its biological significance, its importance in the field of material chemistry can never be underestimated as lots of pyrimidine derivatives are reported recently as constituent of liquid crystal, fluorescence material, photo responsive surfaces etc. attracting the attention of material chemists also. Due to the immense biological, medicinal as well as material significances of pyrimidine derivatives, synthetic chemists have been paying more attention towards development of novel, efficient methodology for various structurally important pyrimidine scaffolds which encourage us to carry out this piece of work.

Chapter 2: A convenient synthesis of novel 5-aryl-pyrido[2,3-*d*]pyrimidines and screening of their preliminary antibacterial properties

This chapter describes a simple methodology for the convenient synthesis of 5-aryl-7-methylpyrido[2,3-*d*]pyrimidines (**3**) by exploiting the diene behaviour of [1-aza-2- (dimethylamino)prop-l-enyl]-1,3-dimethyluracil (**1**). The reaction is

proposed to take place via aza-Diels-Alder pathway followed by elimination of hydrogen molecule, rearrangement and loss of dimethylamine (**Scheme 1**). The synthesized compounds are screened for preliminary antibacterial property and found to have modest activity against both gram positive and gram negative bacteria. Functional modification (-NO_2 to -NH_2) of some selective compounds has been carried out and the functionally transformed products are also exposed to antibacterial screening. Noticeably, functional conversion of the -NO_2 group to -NH_2 leads to enhanced antibacterial activity.

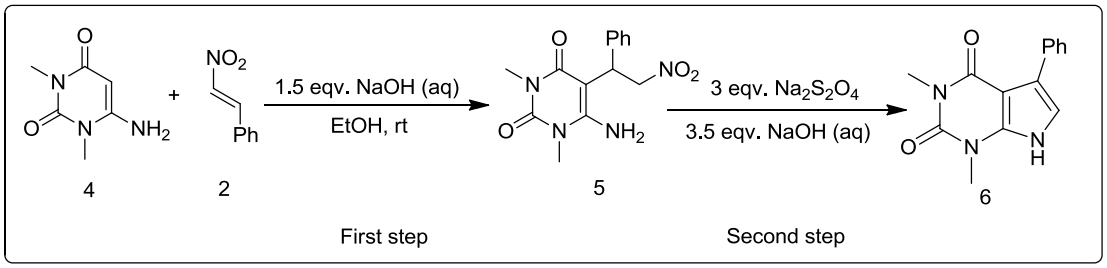


Scheme 1: Plausible mechanism for the formation of 5-aryl-7-methylpyrido[2,3-*d*]pyrimidines

Chapter 3: A one pot, two-step synthesis of 5-arylpyrrolo[2,3-*d*]pyrimidines and their preliminary bioassay studies

This chapter describes a one pot two step methodology for the synthesis of 5-arylpyrrolo[2,3-*d*]pyrimidines (**6**) which involves Michael type addition followed by reductive cyclisation of the adduct. The methodology exploits: (i) the favorability of Michael addition reaction in alkaline medium and (ii) the strong reducing capability of alkaline $Na_2S_2O_4$ solution to carry out the transformation (**Scheme 2**). The methodology is non-catalytic, but still demands attraction as it is quite general for large numbers of nitrostyrenes and possesses comprehensive advantages over most of the earlier methods in terms of reaction time as well as yield. To the best of our knowledge, the described methodology is the first to carry out reductive cyclisation of a molecule possessing an aliphatic nitro group,

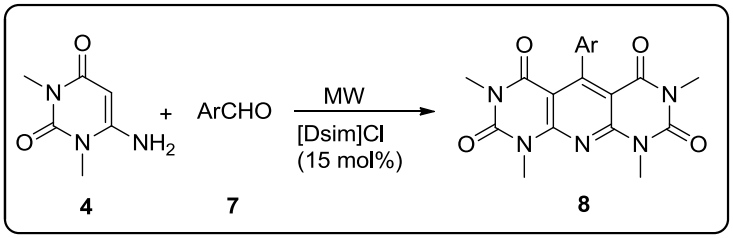
although numerous reports are available for similar conversions in molecules having aromatic nitro group. From biological assay studies, it is found that six of the synthesized compounds show satisfactory activity against *P. aeruginosa* and *B. subtilis*.



Scheme 2: One pot two steps synthesis of 5-arylpyrrolo[2,3-*d*]pyrimidines

Chapter 4: Ionic liquid catalyzed, microwave assisted synthesis of Pyrido[2,3-*d*:6,5-*d*]dipyrimidine-2,4,6,8-tetraones

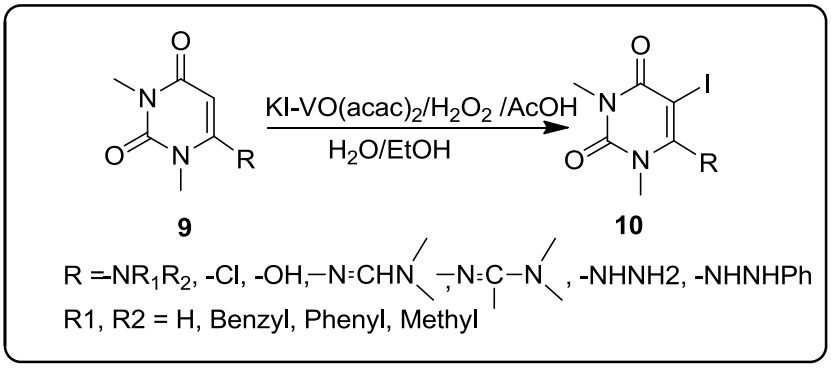
In this chapter, a novel methodology towards the synthesis of pyrido[2,3-*d*:6,5-*d*]dipyrimidine-2,4,6,8-tetrones (**8**) via condensation of 6-amino-1,3-dimethylpyrimidine-2,4(1*H*,3*H*)-dione (**4**) and aldehydes (**7**) using Brønsted acidic ionic liquid as reusable catalyst under microwave irradiation (MWI) has been described (**Scheme 3**). We believe the methodology as the first to carry out the transformation using ionic liquid catalyst coupled with microwave irradiation in absence of added acid.



Scheme 3: [Dsim]Cl catalyzed, microwave assisted synthesis of pyrido[2,3-*d*:6,5-*d'*]dipyrimidine-2,4,6,8-tetraones

Chapter 5: Regioselective iodination at C-5 position of activated pyrimidinediones using KI-VO(acac)$_2$-H$_2$O$_2$-AcOH as iodinating system

This chapter deals with the study towards the development of a convenient method for iodination of activated pyrimidinediones (**9**) using KI-VO(acac)$_2$-H_2O_2-AcOH as a novel iodinating system (**Scheme 4**). Although the methodology is found to be the most effective for regioselective iodination of pyrimidinediones at C-5



Scheme 4: Regioselective iodination of pyrimidinediones

position, it is equally useful for iodination of aromatic amines also. This simple and mild methodology is found to cover a wide spectrum of pyrimidinediones and aromatic amines resulting moderate to excellent yield.

Chapter 6: Concluding remarks and future scopes

This chapter draws an overall conclusion and future scopes of the Ph. D work.