Dihydropyrimidinone chemistry has been actively pursued recently. Close resemblance of 3,4-dihydropyrimidin-2(1H)-ones (DHPMs) with 1,4-dihydropyridines (DHPs), which are potent calcium channel modulators, has led to the breadth and diversity of the subject as reflected by the plethora of published work. Unlike DHPs, dihydropyrimidinones are inherently asymmetric. However, both of these categories of compounds depict identical receptor bound conformation, which has led to the binding site model and identification of conformational preferences depicting interaction with receptor sites, leading to identical calcium channel binding effects. A sizeable number of reports on DHPMs deal with the synthesis and biology, and the others range widely in their focus and scope. The increasing importance of scaffold decoration of heterocycles is attested by the fact that compared to combinatorial chemistry and High Throughput Screening (HTS), a good number of potent drug leads have been rationally designed and synthesized through scaffold decoration approach. The Chapter-2 of the thesis entitled “Review of Literature” amply illustrates the ongoing vitality of the DHPM chemistry, and developments in the classical Biginelli condensation reactions as well as scaffold decoration pathways have been discussed.

Our previous work in this area was mainly focussed on scaffold decoration and we had targeted C-6, C-4, C-5 and N-1 sites to harness diversity in DHPMs. Some of these compounds were also evaluated for their calcium channel binding studies, which led us to develop new synthetic methods to achieve tailor-made target molecules in a synthetically useful manner. To supplement this area of investigation in the present thesis, we have carried out some useful regioselective synthetic transformations (Chapters 3 and 4) on DHPMs, which were either not known or lacked practical utility. Biginelli DHPMs have been converted into highly substituted pyrimidines (Chapter 3), expeditiously, and the pyrimidine derivatives thus obtained have been evaluated as inhibitors of *Mycobacterium tuberculosis* and modulators of cytostatic activity. Compounds were found to possess structure-dependent cytostatic activity. Also we have addressed N-1 and N-3 diversification (Chapter 5) for gaining access to enantiomerically pure DHPMs and have presented satisfactory characteristic data of both the enantiomers, which is extremely scantily reported for this class of molecules.
Using very simple chemical transformation, we have been able to append, organophosphorous as well as phosphorus heterocyclic groups at the N-3 position (Chapter 6). N-3 substitution in DHPMs is important as most of the potent calcium channel blockers among DHPMs bear an acyl, amide or alkyl carboxylate substituents at N-3. Thus, substituting organophosphorous unit at N-3 has added a new category to the existing pool of N-3 substituted DHPMs. While performing N-3 acylation of DHPMs, we obtained facile transformation of N-1,N-3 diacyl DHPMs to N-3 acyl counterparts. Exploiting this opportunity, we have devised a useful synthetic application by performing ‘acyl transfer’ to different nucleophiles during the above transformation. This has led us to describe (Chapter 7) N-1,N-3 diacyl DHPMs as ‘Acyl Transfer Agents.’

Thus, the area of DHPMs offers opportunity to explore synthetic transformations to gain access to structurally modified heterocycles with possible applications in chemistry and biology. Before presenting the results of our investigations, a detailed review on the current status of work in this area is presented in Chapter 2.