Mesoporous MCM-41, one of the promising members of M41S family, has been intensively deliberated with regard to their extensive applications in various tributaries like material science, heterogeneous catalysis, molecular/supramolecular hosts, chemical separation of bulky molecules, biomedicine, membranes and adsorbents due to its peculiar properties.

Chemical modification of MCM-41 by ion exchange is a different approach to tune the catalytic properties. Post synthesis modification of MCM-41 by ion exchange with different metal cations (M) with different Si/M ratio is expected to introduce acidic or basic centres. Hence the detailed study was undertaken in synthesis, characterizations and catalytic properties of MCM-41 materials.

The conclusions drawn from the research study

Novel route of mesoporous MCM-41 synthesis using ultrasonic irradiation and traditional hydrothermal method with various factors influencing the synthesis such as surfactant/silica mole ratios, sources for silica, gel pH, temperature, time etc. have been reported in detail along with crystallization kinetics. It was found that using only ultrasonic irradiation the well developed and highly crystalline phase was obtained within 40 minutes at room temperature. Similarly the hydrothermally synthesised MCM-41 was obtained after 2 hours at 100 °C in basic environment. The detailed comparison between the samples obtained using these two routes of synthesis is studied. It was found that the MCM-41 phase formed using ultrasonic irradiation has higher BET surface area (1185 m²/g) and narrow pore size distribution compared to that of hydrothermally synthesised sample.

It was also found that the pore wall thickness of MCM-41 increases with the power of ultrasonic irradiation. Increasing the wall thickness also enhance the thermal stability of MCM-41. It is an important observations achieved in our study. The observed experimental results indicate that ultrasonic irradiation method is satisfactory for saving time and energy than the hydrothermal synthesis method.

Attempts have been made to find the better source for silica to produce pure MCM-41 with the hydrothermal system. On the basis of time required the silica source reactivity trend was observed

TEOS > Fumed Silica > Sodium Silicate.
Similarly, we also compare the digestive mineralizing agents, it is observed that the aqueous ammonia is more suitable than sodium hydroxide to obtain highly crystalline phase of MCM-41.

In both the ultrasonic and hydrothermal method, concentration of surfactant plays an important role. While we consider the cost effective synthesis, it is necessary to use the less amount of the surfactant. Therefore, we have studied the effect of concentration of surfactant and optimized surfactant/silica ratio to 0.15.

Attempts have been made to study the effect of ultra sound power and irradiation time on the textural properties and morphology of MCM-41. The highly crystalline MCM-41 sample can be synthesized within 40 minutes by ultrasonic irradiation of power 200 W. The synthesized MCM-41 material has larger specific surface area \textit{i.e}.1185 m$^2$/g, pore size 26.8 Å and higher wall thickness \textit{i.e}.18.12 Å.

Chemical modification of well-ordered MCM-41 was carried out by using conventional batch ion exchange technique to obtain the different exchanged forms such as Al-MCM-41, Sn-MCM-41 and Cs-MCM-41. Chemical analysis by EDAX, framework FTIR, SEM, TEM and powder XRD results clearly indicated the exchange of non-framework cations with significant modification in framework composition having no significant modification in phase purity.

From the XRD analysis of all modified samples, one uniform observation witnessed was that the d$_{100}$ values of all the metallosilicates are shifted to higher values compared to MCM-41. This was probably due to the substitution of Si$^{4+}$ by the larger heteroatoms Al$^{3+}$, Sn$^{4+}$ and Cs$^+$ in the framework of MCM-41 and the consequent replacement of the shorter Si-O bonds by the longer M-O bonds causing an expansion of the unit cell ($a_o$). The expansion of unit cell parameter by substitution of Cs$^+$ metal was observed to be more significant as compared to other substituted metal ions \textit{i.e}. Al$^{3+}$ and Sn$^{4+}$. Which was may be due to ionic radius of Cs$^+$ ion is greater than the Al$^{3+}$ and Sn$^{4+}$ metal ions. The change in the M-O-M bond angle on heteroatom substitution probably causes some long-range disorder in the system leading generally to slightly broader XRD lines for the metallosilicates compared to the parent MCM-41. Also, there are marginal changes in the crystallinity but almost no changes in the phase purity and structural morphology are being observed after modification with different Si/M ratio which implies the structural stability of the parent sample and presence of metal ions in the framework of MCM-41.
The UV-Vis analysis revealed that most of the exchange cations occupy the framework position. When we compare the EDAX data obtained for these three guest atoms, it was identified that at the same concentration of exchange solution, the Si/M ratio (where, M = Al$^{3+}$, Sn$^{4+}$ or Cs$^+$) increases with increase in the cationic size. UV-Vis spectra analysis shows a characteristic absorption band around 208 nm in case of Sn-MCM-41 samples. The intensity of this absorption band increases with increasing Sn content which concludes the presence of Sn in tetrahedral coordination in the walls of MCM-41 made up of Si$^{4+}$ ions.

The attempts have been made to study the hydrating ability for the higher degree of exchange samples of MCM-41 on the basis of thermal analysis. The hydrophobicity trend of MCM-41 along with its exchanged forms on the basis of weight loss during thermal analysis is as follows:

MCM-41 > Cs-MCM-41 > Sn-MCM-41 > Al-MCM-41

Thus, here we have proved that the surface silanol groups are responsible for hydrophilicity of mesoporous MCM-41 materials. Above same trend was observed for ΔH, obtained from DSC analysis.

When we consider the degree of exchange of Al cations the hydrophilicity increases with increase in the Al content of MCM-41. This was also confirmed by FTIR study.

Synthesised parent MCM-41 and its exchange forms Cs-MCM-41 were catalytically evaluated for the Knoevenagel condensation reaction. The performance of Cs exchange samples depends on degree of exchange, hence the highest exchanged sample showed high conversion and nearly 100 % selectivity. This may be related to the low electronegativity nature of Cs, which increases the negative charge on the framework oxygen leading to higher basicity of MCM-41.

Another studied condensation reaction was multicomponent Biginelli reaction over siliceous MCM-41. It was observed and concluded that MCM-41 can serve as an efficient catalyst for the synthesis of DHPMs. Also followed reaction procedure offers several advantages including mild reaction conditions, cleaner route, and high yields of product. This simple procedure combined with easy recovery of catalyst and reuses of the catalyst makes this method economic, benign, and a waste-free chemical process for the synthesis of these compounds. We believe that this procedure is appropriate, cost-effective, and comprehensible process for the synthesis of 5-acetyl-4-phenyl-6-methyl-3, 4-dihydropyrimidine-2-(1H)-one.
An eco-friendly and benign catalytic process, employing solid acid catalysts for acylation of benzene to give acetophenone is presented in Chapter 4B. Al-MCM-41 has replaced traditional catalyst in Friedel Craft acylation reaction which was hazardous and produces stoichiometric amount by-product.

On the basis of conversion of acetyl chloride, the activity of catalyst follows the order Al-MCM-41 (20) > Al-MCM-41 (35) > Al-MCM-41(52) and this is the order of density of acidic sites of Al-MCM-41 samples obtained from NH₃-TPD. Therefore, it is found that both acidity and conversion increase with Al content. Also it is found that catalyst can be recycled up to 5 times. While studying the effect of catalyst loading, observations and interpretation of the result suffices the need for lower amount of catalyst i.e. 0.4 g to carry out the reaction. Thus, the Al-MCM-41 appears to be an auspicious catalyst for the Friedel-Crafts acylation of benzene.

The Sn and Al incorporated MCM-41 metal catalysts are studied for the liquid phase esterification of acetic acid by the n-butyl alcohol to give n-butyl acetate. The effects of different reaction parameters such temperature, alcohol to acetic acid mole ratio, catalyst concentration and metal content are reported for Al-MCM-41 catalysts. The activities of Sn- and Al- incorporated MCM-41 catalysts, were compared at similar reaction conditions. Al-MCM-41 is found to be the most active catalyst for esterification of acetic acid.

The obtained data of NH3-TPD revealed that as Sn-MCM-41 (22) has nearly same acidity as compared to Al-MCM-41(35) so that we except the % conversion should be nearly 72 % but obtained results are not in line with our view. Though Sn-MCM-41 samples possess sufficient acidity it cannot accelerate esterification reaction. This was may be due to catalysts Sn-MCM-41 contains more Lewis acidic sites than the Brönsted acid sites. Thus we have conclude that esterification of acetic acid with n-butyl alcohol is catalysed by strong Brönsted acid sites. Also in our research study we have investigated that in Sn-MCM-41 materials the acidity is presumably of the Lewis type and arises from Sn (IV) cations. As Sn is present in the catalyst as Sn⁴⁺, it is unlikely that Brönsted acidity is created, as when trivalent ions are introduced.