Summary

This work is an effort to probe into the supramolecular assemblies formed by low molecular weight compounds and their ability to exhibit gelation behavior. These gels were also explored for various potential applications.

- A series of 12 salts of memantine were designed and synthesized using combinatorial library approach. It was found that most of the structures displayed supramolecular synthons with graph set representation, $R_4^2(8)$ and $R_4^4(12)$.
- Memantine (MEM) has a great therapeutic utility as it is utilized in the treatment of several forms of dementia, like the Alzeimer's, Parkinson's and Huntington's diseases.
- While no gels could be obtained in this series of salts, the newly synthesized salts showed decent solubility behavior and a strong dependence on van der Waals interactions. We believe that the hydrochloride salt can be replaced by some organic salt with almost comparable solubility values and we can consider this study as a promising one for development of memantine formulations in future.
- Pivalic acid based salts were synthesized on the working hypothesis that 1D H-bonded network supports gelation; out of which, ten salts turned out to be organogelators.
- Even though the crystal engineering approach to design a new gelator proved to be fruitful, it could not provide a complete understanding of supramolecular gelation due to lack of information about the role of solvents on gelation behavior.
- The role of solvent parameters in controlling gelation was also established, where it was found that bulk solvent properties of a solvent (Dielectric constant- $\boldsymbol{\varepsilon}$, and polarity index- ***P'***) may indicate its propensity to gel a solvent. The Kamlet-Taft hydrogen bond accepting parameter ($\boldsymbol{\beta}$) and polar cohesive energy parameter ($\boldsymbol{\delta_p}$) displayed a positive correlation with the thermal stability of gel. The mgc values were found to be positively related to the $\boldsymbol{\delta_h}$ values, the hydrogen bonding HSP.
- A series of *tert*-butylacetic acid based salts was synthesized to probe into the gelation performance and compare it with that of pivalic acid based salts. We believe that an additional methylene group fails to attain the delicate balance between hydrophobic and hydrophilic forces required for exhibiting gelation behavior.

- The gelation performance was found to be compromised in comparison with that of pivalic acid salts. Gels were mainly formed in DMSO and DMF at much higher mgc values than the pivalic acid salts with much lesser T_{gel} values.
- A group of pivalic acid based amides were synthesized based on **π/Al-A-B** (aromatic/aliphatic-amide linker-bulky group) type design. Six new gelators were discovered which were extensively exploited to showcase an exceptional gelation behavior towards aliphatic long-chain solvents in various conditions (viz. external heat, strong sunlight, UV light, and ultrasonication). By far, these compounds are some of the smallest molecules (prepared by easy one-step synthesis) employed for successful phase selective organogelation in water-oil type mixtures, with an extraordinary gelation performance in both powder form and presence of secondary solvents, offering a viable method for easy recovery and good recyclability, also applicable in a typical environmental setting.
- A similar library of *tert*-butyl acetic acid based amides were also synthesized to assess and modify the gelation behavior in alkane homologues and commercial fuels. However, it was found that the solubility of these compounds was found to be sluggish, as the gels could be formed only when the system was heated upto high temperature (65-70°C). No gelation was observed in presence of strong sunlight. A UV irradiation for atleast 15 minutes or ultrasonication for 15-20 minutes at 50°C was required to induce gelation. These compounds were also tested for their ability to bind with heavy metal ions in aqueous medium with superior detection limits.
- Both these sets of compounds were exploited to absorb dyes from aqueous medium with impressive removal efficiencies.
- Pivalamides containing heterocyclic backbones were synthesized and utilized for synthesized copper(II) metal complexes. These complexes were characterized using single crystal X-ray studies and computational studies. These were also employed to assess their superoxide dismutase (SOD) mimic activity, BSA and ct-DNA binding abilities. The IC_{50} values for the complexes obtained in the SOD mimic studies were found to be much lower than those reported so far in the literature. DNA cleavage studies were also performed using gel electrophoresis.