Title:	Harnessing Fluid Actuation & Liquid-in-Liquid Sculpting using Supramolecular Constructs.
Authors:	Agashe, Chinmayee Uday.
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Abstract:	Supramolecular chemistry has been described as 'chemistry beyond the molecule' as it focuses on designing and structuring functional chemical systems by assembling molecular components through noncovalent interactions. Supramolecular constructs can be fabricated by appropriate manipulation of weak noncovalent intermolecular forces such as hydrogen bonding, electrostatic interactions, van der Waals forces, hydrophobic interactions, host-guest interactions, pi stacking as well as stronger dynamic covalent bonds. Owing to their unique properties, supramolecular constructs have a wide range of applications including drug delivery, sensing, catalysis, bioimaging, environmental remediation, fabrication of molecular machines and microscale devices etc. Over the recent years, there has been significant interest to mimic complex natural motions in the synthetic realm. In pursuit of such smart systems, supramolecular constructs provide a promising tool. For fabrication of microscale devices with fluid actuation, there have been several reports of micropumps which can work without an aid of external power source in response to external stimuli. A chemically powered pump can enable fluid actuation in presence of specific analytes based on the complementarity. Supramolecular micropumps based on host-guest interactions have been shown to devise powerless microfluidic devices where fluid flow can be manipulated by modulating the noncovalent interactions. In such cases, the 'host' molecule with aesthetic architecture of intramolecular cavity can noncovalently interact with the 'guest' molecule and trigger fluid flow. Utilizing this concept, in first work we fabricated a non-mechanical, self-powered pillar[5]arene multilayer film based micropump to establish flow-based detection of micropollutants. The host-guest interaction between pillar[5]arene and micropollutants turned on fluid flow inside the micropump chamber. We also demonstrate the comparison of fluid pumping velocity of 2 model micropollutants – Bisphenol-A and 2-Naphthol based on thermodynamic as well as numerical modelling. The fluid pumping velocity showed dependency on the concentration of micropollutant solution as well as the no. of bilayers of the host multilayer film. In second work, we utilised another host molecule cucurbit[7]uril to modulate enzyme catalysis and subsequently regulated fluid flow in an enzyme micropump. In this work, bovine carbonic anhydrase (BCA) multilayer film was coated onto substrate and enzyme micropump was fabricated using 4-Nitrophenyl acetate as the substrate. A two-faced guest molecule containing BCA binding epitope and cucurbit[7]uril binding epitope was synthesized and utilised to inhibit the enzyme. The inhibition of enzyme xiiiwas reversed by capture of guest molecule by cucurbit[7]uril thereby restoring the activity of the enzyme and fluid motion in the micropump. Due to surge in demand of soft and liquid based devices for practical applications, liquids with non-equilibrium shapes and thus the concept of liquid sculpting have garnered substantial attention in the past decade. In case of all liquid systems, when components of one phase adsorb onto the interface and react with the components of the adjacent phase, the overall binding energy increases. A system of supramolecular constructs provides a substantial means to generate such interfacial assemblies. When a compressive force is applied to such an interface, jamming occurs and interface buckles. This can be employed to fabricate nature mimicking compartments. Using this approach, in our third work we utilised dynamic imine covalent bonding under the umbrella of supramolecular chemistry to form anisotropic compartments at the liquid-liquid interface. In this work, the reaction between water soluble polyethyleneimine and oil soluble aldehydes rendered anisotropic compartments which can be used to trigger stimuli responsive cargo release. In our fourth work, we utilized pi stacking of perylenediimide fluorophore to show AIE-switching and chiral recognition of basic amino acids. In summary, this thesis emphasizes on employing uniqueness of supramolecular constructs to initiate and regulate fluid flow in microscale devices and sculpting of liquids for fabricating dynamic and responsive interfaces.
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