

Guest Editorial: Responsive Host–Guest Systems

The term host–guest chemistry was first introduced by Donald Cram in 1976 to describe the chemistry of complexes formed by two or more molecules or ions that are held together in certain structures through noncovalent interactions. *Host is defined as an organic molecule or ion whose binding sites converge in the complex, while guest is defined as any molecule or ion whose binding sites diverge in the complex.* There are several generations of hosts in host–guest chemistry, from crown ethers and cryptands to calixarenes and pillararenes. Host–guest systems that can respond to external stimuli have attracted much attention due to their applications in the fields of chemosensors, supramolecular polymers, drug delivery, and biological imaging. Many interesting host–guest systems, such as molecular elevators, molecular muscles, molecular necklaces, and molecular locks, have been constructed. At the same time, there has been rapid progress in understanding the physiochemical processes of host–guest complexation, the relative movements in host–guest complexes triggered by different stimuli, the bioactivity and biocompatibility of the hosts, and the association and disassociation in the formation of supramolecular polymers. The purpose of this special issue on “Responsive Host–Guest Systems” is to collect a series of Accounts by the leading experts in supramolecular chemistry to provide an overview of recent major developments in these respects. From this special issue, the readers can quickly learn how to construct highly ordered and advanced functional systems based on host–guest recognition. These Accounts can be divided into five thematic groups based on the emphasis of each Account.

The first group of three Accounts discusses progress based on two of the most venerable supramolecular hosts: Isaacs and Kaifer focus on functional systems based on cucurbiturils. Liu reviews the unique recognition and assembly properties and potential biological applications of a type of water-soluble calixarene, *p*-sulfonatocalix[*n*]arene.

The second group of eight Accounts addresses the development of new types of supramolecular hosts based on the authors' efforts. Liu, Helgeson, and Houk provide experimental and computational investigations of gated hemi-carcerands and several potential applications of gated hemi-carceplexes based on Cram's resorcinarenes. Huang and Gibson discuss how to design crown ether-based cryptands and how these cryptands can be used to increase association constants with organic guests and prepare functional supramolecular systems efficiently. Chen focuses on iptycene-derived crown ether hosts. Nitschke summarizes stereochemistry in metal–ligand complexes based on subcomponent self-assembly. Li discusses how aromatic amide and hydrazide foldamers induced by hydrogen bonding can be used to produce responsive host–guest systems. Shimizu highlights columnar assembly in high fidelity based on bis-urea macrocycles constructed from two C-shaped spacers and two urea groups. Diederich presents the development of molecular grippers from redox-switchable resorcin[4]arene cavitands. Aprahamian talks about hydration-based molecular switches.

Seven Accounts constitute the third group that centers on the construction of stimuli-responsive supramolecular complexes. Stoddart summarizes rotaxane-based molecular muscles. Wang focuses on dynamic supramolecular complexes based on orthogonal self-assembly. Tang talks about drug and gene delivery systems constructed by host–guest molecular recognition. Wu and Tung review enhancement of the efficiency of photocatalytic reduction of protons to hydrogen via molecular assembly. Zhang and Scherman presents multifunctional biointerfaces fabricated by host–guest interactions and supramolecular chemistry at interfaces, while Yang addresses switchable host–guest systems on surfaces. Thayumanavan talks about supramolecular disassembly of facially amphiphilic dendrimer assemblies in response to physical, chemical, and biological stimuli.

The fourth group of six Accounts mainly focuses on supramolecular polymers and supramolecular gels formed by macrocycle-based host–guest interactions. Harada reviews supramolecular polymeric materials through cyclodextrin–guest interactions. Huang summarizes state-of-the-art progress in the area of supramolecular polymers based on five types of classical supramolecular hosts. Tian focuses on the stimuli-responsive supramolecular polymers based on cyclodextrins, cucurbiturils, and hydrophilic calixarenes in aqueous solution. Zhu gives insight on the syntheses, functionalization, self-assembly, and potential applications of supramolecular dendritic polymers. Liu presents progress in responsive polymeric materials based on host–guest recognition motifs and highlights their applications in optical sensing and imaging, drug and gene delivery, and self-healing materials. Schalley addresses the construction of supramolecular gels based on macrocycles.

The last group of four Accounts showcases recent achievements in the development of supramolecular sensors. Anslin develops rapid optical methods for enantiomeric excess analysis. Beer summarizes sensing of charged guest species by rotaxanes and catenanes. Nau gives insight on analyte-responsive macrocyclic host–fluorophore systems, while Miljanić presents fluorescent sensors based on benzobisoxazole cruciform.

We are very grateful to all authors involved for their contributions to these wonderful Accounts contained in this special issue of *Accounts of Chemical Research*. The development of responsive host–guest systems and their potential applications is a multidisciplinary endeavor, one that demands input from scientists with various backgrounds. We are sure that this special issue gives the readers insights into the world of research on responsive host–guest systems, and we hope that these Accounts will convince young scientists to devote new creative efforts in this area.

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Notes

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