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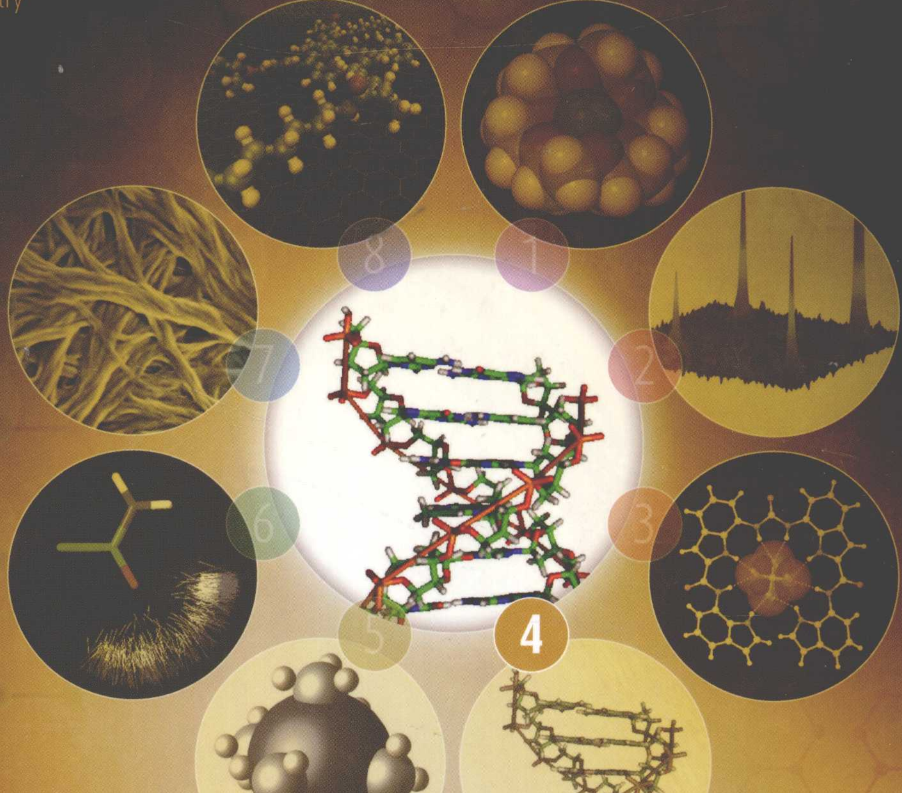

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Supramolecular Chemistry: From Molecules to Nanomaterials

Volume 4: Supramolecular Catalysis, Reactivity and Chemical Biology

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Foreword

Supramolecular chemistry has been defined as "chemistry beyond the molecule". It aims at constructing highly complex, functional chemical systems from components held together by intermolecular noncovalent forces. It has relied on the development of preorganized molecular receptors for effecting molecular recognition, on the basis of the molecular information stored in the covalent framework of the components and read out at the supramolecular level through specific interactional algorithms. Suitably functionalized receptors may display supramolecular reactivity and catalysis and selective transport processes.

A most basic and far-reaching contribution of supramolecular chemistry to chemical sciences has been the implementation of the concept of molecular information. It involved the storage of information at the molecular level; in the structural features; and its retrieval, transfer, and processing at the supramolecular level, through molecular recognition processes operating via specific spatial relationships and interaction patterns. Supramolecular chemistry has thus paved the way toward apprehending chemistry also as an information science.

The control provided by recognition processes allowed the development of functional molecular and supramolecular devices, defined as structurally organized and functionally integrated systems built from suitably designed molecular components performing a given action (e.g., photoactive, electroactive, and ionoactive) and endowed with the structural features required for assembly into an organized supramolecular architecture. Thus emerged the areas of supramolecular photonics, electronics, and ionics.

Beyond mastering preorganization and taking advantage of it, supramolecular chemistry has been actively exploring the design of systems undergoing self-organization, that is systems capable of spontaneously generating well-defined, organized supramolecular architectures by self-assembly from their components, under the control of molecular information processes. They operate as programed chemical systems and are of major interest for supramolecular science and engineering. They give access to advanced functional supramolecular materials, such as supramolecular polymers, liquid crystals and lipid vesicles as well as solid-state assemblies.

The implementation of 'programed' self-organizing systems amounts to performing self-organization by design.

It also provides an original approach to nanoscience and nanotechnology. In particular, the generation of well-defined, functional supramolecular architectures of nanometric size through self-organization represents a means of performing programed engineering and processing of nanomaterials. Technologies resorting to self-organization processes are, in principle, able to provide a powerful complement and/or alternative to nanofabrication and nanomanipulation procedures by making use of the spontaneous but controlled generation of the desired superstructures and devices from suitably instructed and functional building blocks. The long-range goal is to shift from entities that need to be made to entities that make themselves, that is, from fabrication to self-fabrication.

From another point of view, self-organization is, in principle, able to select the correct molecular components for the generation of a given supramolecular entity from a diverse collection of building blocks. It may thus take place with selection, by virtue of a basic feature inherent in supramolecular chemistry, that is, its dynamic character.

Indeed, supramolecular chemistry is intrinsically a dynamic chemistry in view of the lability of the interactions connecting the molecular components of a supramolecular entity and the resulting ability of supramolecular species to exchange their constituents. Such a dynamic character is also conferred to molecular chemistry when the molecular entity contains covalent bonds that may form and break reversibility, so as to allow a continuous change in constitution by reorganization and exchange of building blocks. Thus, supramolecular chemistry has also fertilized molecular chemistry, leading to the definition of a Constitutional Dynamic Chemistry on both the molecular and supramolecular levels. It takes advantage of dynamic diversity to allow variation and selection. It operates on dynamic constitutional diversity in response to either internal or external factors to achieve adaptation.

Supramolecular chemistry has progressed over the years along three overlapping phases. The first is that of molecular recognition and its corollaries, supramolecular reactivity, catalysis, and transport; it relies on design and preorganization and implements information storage and processing.

The second concerns self-assembly and self-organization, that is self-processes in general; it relies on design and implements programing and programed systems for controlling the generation of specific entities in complex mixtures.

The third concerns constitutional dynamics of both molecular and supramolecular entities, defining a constitutional dynamic chemistry as a unifying concept. It relies on self-organization with selection in addition to design, and leads to the emergence of adaptive and evolutive chemistry.

Since it has been named in 1978 by the undersigned, about 10 years after the seed had been planted, the field of supramolecular chemistry has experienced a spectacular growth at the triple meeting point of chemistry with biology and physics. Its concepts and the perspectives it opens have been delineated, attracting scientists with a wide range of expertise. It has given rise to numerous review articles and special issues of journals and books. The present monumental work comes very timely. It provides thorough reviews and discussions, covering a broad range of topics, authored by many of the major players in the field. It takes stake and opens perspectives to the creative imagination of all participants in our common adventure.

I would like to very warmly congratulate and thank the editors and the contributors alike for this precious gift to the science of chemistry.

Jean-Marie Lehn
July 2011

Preface

Over the past decade, there have been tremendous advances in our understanding of the way in which chemical concepts at the molecular level build up into materials and systems with fascinating, emergent properties on the nanoscale. Creating that link between the chemist's understanding of the way in which molecules interact with one another and the understanding a materials scientist, engineer, or biologist has of the resulting properties of a material or system composed of those molecules is one of the huge, grand challenges facing modern molecular science. This vision of a molecular-level approach to complex systems and materials is the underlying drive for this project.

In 1996, the impressive *Comprehensive Supramolecular Chemistry* was published. This substantial 11-volume work summarized all of the major systems studied in fields based in supramolecular chemistry since its inception in clathrate chemistry in the early nineteenth century and cation receptor chemistry in the mid-1960s. In the 15 years since, the field has blossomed enormously and supramolecular concepts have become much more integrated into modern science underlying many areas that are based, fundamentally, on molecules. In attempting to capture and catalyze that continuing development, we have adopted a very different vision for this project. We aim to produce an enmeshed overview of the concepts and techniques of modern supramolecular chemistry and show, based on fluent chapters by the leading international experts, how these paradigms evolve seamlessly into nanoscale systems chemistry and materials science, and of course beyond. The scope and coverage has been carefully designed by the Editorial Advisory Board and the Editors of the 10 sections to avoid mere summative descriptions and instead to produce an interlocking series of tutorial-style articles that guide advanced students and veteran practitioners swiftly to the key science and techniques used in addressing modern supramolecular and nanoscale chemistry. We and the Board have taken particular care to try to break down the barriers between synthetic chemistry and materials science and show how modern techniques allow access increasingly far along the "synthesising up" pathway. We hope that this conceptual basis and forward-looking narrative is useful and complements the fascinating descriptions of earlier work published in 1996.

The origins of this work lie in a very successful "fun day" of science organised by Thorri Gunnlaugsson at the Trinity College, Dublin, in 2008, and we thank Thorri for being the initial catalyst and such a tremendous host. A large subsection of the editorial and advisory boards met at the *International Symposium for Supramolecular and Macrocyclic Chemistry* at Maastricht, Netherlands, in 2009, and between them defined the scope and structure of the project. The 10 editors then translated these concepts into a detailed vision for their own sections. We are hugely grateful to everyone on the advisory and editorial boards who gave their time, energy, and reputations to this project. Their belief has been invaluable, and as editors-in-chief, we feel that the result has vindicated their commitment. Our greatest debt goes, of course, to the authors themselves who have had the hugely challenging task of translating this concept-based vision into reality in the 169 individual chapters, each one a significant scientific product in its own right. We feel they have done an excellent job and salute their fidelity to the project's values.

We would also like to express our tremendous gratitude to Paul Deards at Wiley who believed in us all through this idea and brought it to reality. The project would also have come to nothing without the mountain-moving organizational skills of Stacey Woods and Anne Hunt at Wiley who have worked tirelessly to keep the momentum moving forward and "herd cats" as well as bring the book to the standard and accessibility it needs to have. J. W. S. is very grateful to the Durham University for providing two terms of research leave, which made this project and the travel it needed much easier to achieve, and we are both as ever indebted to the many fine coworkers who have passed through our laboratories over the years who make chemistry such an enjoyable subject to work in. P. A. G. thanks Nittaya for her love and support. J. W. S. would like to offer an ongoing thanks to his partner Kirsty, an ever-present source of wisdom and voice of sense.

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Abbreviations and Acronyms

17-AAG	17-Allylamino-17-demethoxygeldanamycin
2-AP	2-Aminopyrimidine
9-ap	Anthracene-9-propionic Acid
4-bn-res	4-Benzylresorcinol
4,4′-bpe	*Trans*-1,2-bis(4-Pyridyl)Ethylene
bpea	1,4-bis(4-Pyridyl)Ethane
1,4-bpeb	1,4-bis[2-(4-Pyridyl)Ethenyl] benzene
bpee	1,4-bis(4-Pyridyl)Ethene
1,4-bpef	*p*-Di-[2-(4-Pyridyl)Ethenyl]-2-fluorobenzene
bpp	1,3-bis(4-Pyridyl)Propane
1,5-bppo	Bis(4-Pyridyl)-1,4-pentadiene-3-one
2-CH_3THF	2-Methyltetrahydrofuran
4-Cl dpcb	*Rctt*-1,2-bis(4-Pyridyl)-3,4-bis (*p*-Chlorophenyl)Cyclobutane
4-Cl stilbz	4-Chlorostilbazole
1D	One-Dimensional
2D	Two-Dimensional
3D	Three-Dimensional
9EA	9-Ethyladenine
6HB	Six-Helix Bundle
8HB	Eight-Helix Bundle
2,3-nap	2,3-bis(4-Methylenethiopyridyl)-Naphthalene
1,8-nda	1,8-naphthalenedicarboxylic Acid
4-pa	(*E*)-3-(4-Pyridyl)Acrylic Acid
6PE	Sixfold Phenyl Embraces
6PGL	6-Phosphogluconoylation
4-py-but	*Trans*-1,4-(4-Pyridyl)-1,3-butadiene
4-py-hex	*Trans*-1,6-(4-Pyridyl)-1,3,5-hexatriene
4-vp	4-Vinylpyridine
AAO	Anodized Aluminum Oxide
ABC	Adenosine-5′-triphosphate Binding Cassette
ABZ	Albendazole
AC	Alternating Current
ACA	Acetoxychavicol Acetate
acac	Acetylacetonate
ACE	Affinity Capillary Electrophoresis or Angiotensin Converting Enzyme
ACh	Acetylcholine
ACHC	Aminocyclohexanecarboxylic Acid
AChE	Acetylcholine Esterase
AcOH	Acetic Acid
ACR	Aza-Crown Resorcinarene
aCTG	Triamino Cyclotriguaiacylene
ACU	Undecyl-Aza-18-crown-6
AD	Acceptor Dendrimers or Activating Domain
Ad	Adamantane
Ad-PEG	Ad-Modified Polyethylene Glycol
ADA	Acceptor Donor Acceptor
ADMET	Acyclic Diene Metathesis
ADP	Adenosine 5′-Diphosphate
ADR	Adriamycin
ADV	Adenovirus
aeg	*N*-(2-Aminoethyl)-Glycine
AEM	Arylene Ethynylene Macrocycle
AFM	Atomic Force Microscopy
AFP	Alpha-Fetoprotein
AgNP	Silver Nanoparticle
AHX	ε-aminohexanoic Acid
AIBN	2,2′-Azobis(Isobutyronitrile)
AIEE	Aggregation-Induced Enhanced Emission
AK	Attenuated *K*
ALD	Atomic Layer Deposition
ALK	Ala-Leu-Lys-Arg-Gln-Gly-Arg-Thr-Leu-Tyr-Gly-Phe
ALP	Alkaline Phosphatase
ALP	Amphiphilic Lipopeptide
AM1	Austin Model 1
AM1.5	Air Mass 1.5
AMF	Alternating Magnetic Field
AMFE	Anomalous Mole Fraction Effect
amm-stilb	Bis(Dialkylammonium)-Substituted Stilbene
AMP	Adenosine 5′-Monophosphate
AMT	Amitriptyline Hydrochloride
ANB	5-Azido-2-nitrobenzoic Acid Chloride
ANN	Artificial Neural Network

ANTS	8-Aminonaphthalene-1,3,6-trisulfonate
AO	Atomic Orbital
AP	Aptamer–Photosensitizer
APC	2,4-bis(4-Dialkylaminophenyl)-3-Hydroxy-4-alkylsulfanylcyclobut-2-enone
APED	Alternating Polyelectrolyte Deposition
API	Active Pharmaceutical Ingredient
aPP	Avian Pancreatic Polypeptide
APTES	Aminopropyltriethoxysilane
AQ	Anthraquinone
AR	Aromatic Resin
AR	Aviram-Ratner
ASGPR	Asialoglycoprotein Receptor
Atb	*S*-2-Amino-4-trifluorobutyric Acid
ATCh	Acetylthiocholine
ATP	Adenosine 5′-Triphosphate
ATR-FTIRS	Attenuated Total Reflection-Fourier Transform Infrared Spectroscopy
ATR-IR	Attenuated Total Reflectance Infrared
ATRP	Atom Transfer Radical Polymerization
AU	Analytical Ultracentrifugation
Au-SNP	Au Supramolecular Nanoparticle
AUC	Analytical Ultracentrifugation
AuNP	Gold Nanoparticle
AV	Ala-Val
AZTDP	Azido-3′-deoxythymidene 5′-diphosphate
AZTMP	3′-azido-3′-deoxythymidene 5′-monophosphate
BAM	Brewster Angle Microscopy
BAMP-ligand	Bis(aminomethyl)pyridine Ligand
BAPTA	1,2-bis(o-aminophenoxy)ethane-*N,N,N′,N′*-tetraacetic acid
BAR	Barbituric Acid
BASE	Boron Affinity Saccharide Electrophoresis
BASF	Baden Aniline and Soda Factory
BBV	Boronic Acid-Substituted Benzylviologen
BCA	Bio-bar-code amplification
BCB	Benzocyclobutane
BCC	Body Centered Cubic
BCD	β-Cyclodextrin
BCP	Block Copolymer
bdc	1,4-benzenedicarboxylate
BDC	Benzenedicarboxylic Acid
BDE	Bond-Dissociation Energies
BDG	Benzodiguanamine
bdta	1,2,4,5-benzenetetracarboxylic Acid
BET	Brunauer Emmett Teller or Back Electron Transfer
BFDMA	Bis-(11-Ferrocenylundecyl)-Dimethylammonium Bromide
bFGF	Basic Fibroblast Growth Factor
BH	Bcl-2 Homology
BI	Bovine Insulin
BIA	Biomolecular Interaction Analysis
BIC	5-(Benzyloxy)-Isophthalic Acid
BINAM	2,2′-diamino-1,1′-binaphthalene
BINAP	2,2′-bis(Diphenylphosphino)-1,1′-binaphthyl
bipy	Bipyridine
BiTE	Bispecific T-Cell Engager Molecules
bix	1,4-bis(Imidazol-1-ylmethyl)-Benzene
BLM	"Black" Lipid Membrane
BM	Ball Milling
BMP	Bone Morphogenetic Protein
BNCT	Boron Neutron Capture Therapy
BNP	Binaphthyl Phosphate
BNPP	Bis(4-Nitrophenyl)Phosphate
BO	Butylene Oxide
BODIPY	Boron Dipyrromethane
BoNT	*Botulinum Neurotoxin*
BP	Biphenol
BPA	Bipyridine Amine
BPB	Bromophenol Blue
BPEA	Bis-(Phenylethynyl)Anthracene
bPP	Bovine Pancreatic Polypeptide
BPP34C10	Bis(*p*-Phenylene)-34-Crown-10
bpp-34-crown-10	Bisparaphenylene-34-crown-10
BPPM	*N*-tert-butoxycarbonyl-4-diphenylphosphino-2-diphenylphosphino-mehtyl-pyrrolidine
bpy	Bipyridine
BRGD-PA	Aspartate-Arg-Gly-Asp
BSA	Bovine Serum Albumin
BSM	Bovine Submaxillary Mucin
BSP	Bone Sialoprotein
BTA	Benzene-1,3,5-tricarboxamide
BTB	1,3,5-benzenetribenzoate
BTC	1,3,5-benzenetricarboxylic Acid
BTC	Benzene-1,3,5-tricarboxylate
BTE	Backbone Thioester Exchange
BTF6	1,2-bis(2-Methylbenzo[b]thiophen-3-yl)Hexafluorocyclopentene
BTM	Benzotetramisole
BTMA	*n*-Butyltrimethylammonium
BTX	Bent Triple-Crossover
BZ	Belousov–Zhabotinsky
BZD	Benzidine

CA	Carbonic Anhydrase or Cyanuric Acid
CAC	Critical Aggregation Concentration
CAHBs	Charge-assisted H-bonds
cAMP	Cyclic Adenosine Monophosphate
CAP	Chloramphenicol
CAP-MR	Chloramphenicol-methyl Red
CAS	Chrome Azurol S
CB or CB[*n*]	Curcurbit[*n*]uril
CB[6]	Cucurbit[6]uril
CB[7]	Cucurbit[7]uril
CB[8]	Cucurbit[8]uril
CBA	4-Carboxyphenylboronic Acid
CBED	Convergent-Beam Electron Diffraction
CBPQT^{4+}	Cyclobis(Paraquat-*p*-phenylene)
cbta	Cyclobutanetetracarboxylic Acid
CC	Coupled Cluster
CCA	Colloidal Crystalline Array
CCD	Charge Coupled Device
CCDC	Cambridge Crystallographic Data Centre
ccdc	Cobaltocenium-1,1′-dicarboxylate
CCK8	Cholecystokinin Octapeptide
CCMV	Cowpea Chlorotic Mottle Virus
ccnm	Carbamoylcyanonitrosomethanide
ccp	Cubic Close-packed
CCW	Counterclockwise
CD	Circular Dichroism or Cyclodextrin
α-CD	α-Cyclodextrin
β-CD	β-Cyclodextrin
γ-CD	γ-Cyclodextrin
CD-PEI	Cyclodextrin-Modified Polyethyleni-mine
CD/Ad	Cyclodextrin/adamantine
CDCs	Cholesterol-Dependent Cytolysins
CDI	1-(3-Dimethylaminopropyl)-3-Ethylcarbodiimide
CDI	Coherent Diffraction Imaging
cdo	Diolefin Chelidonic Acid
CDP	Cyclodextrin-Based Polymer
CdSe	Cadmium Selenide
CDV	Cyclodextrin Vesicle
CE	Capillary Electrophoresis
CEC	Capillary Electrochromatography
CEST	Chemical Exchange Saturation Transfer
CF	5(6)-Carboxyfluorescein
CFET	Chemical-Field-Effect Transistor
CFSE	Crystal Field Stabilization Energy
CGOM	Crystal Growth of Organic Materials
CHAPS	3-[(3-Cholamidopropyl)Dimethyl-ammonio]-1-propanesulfonate
CHEF	Chelation-Enhanced Fluorescence
CHEMFET	Chemically Modified Field-Effect Transistor
CHO	Chinese Hamster Ovarian
CH$_3$OH	Methanol
CHO-K1	Chinese Hamster Ovary
ChS	Chondroitin 4-Sulfate
CHTE	Cyclohepta-1,2,4,6-tetraene
CI	Configuration Interaction
CID	Collision-Induced Dissociation
C-IDA	Colorimetric Indicator Displacement Assay
CIF	Crystallographic Information File
CIGS	Cuin$_x$Ga$_{(1-x)}$Se$_2$
CK II	Casein Kinase II
CL	Chemiluminescence
CLC	Cholesteric or Columnar Liquid-Crystalline
CLIO	Crosslinked Iron Oxide
CLs	Chemical Leitmotifs
CLSM	Confocal Laser Scanning Micro-scopy
ClSubPc	Chlorosubphthalocyanine
CMC	Critical Micellar Concentration
CME	Chemically Modified Electrode
CMOS	Complementary Metal Oxide Semicon-ductor
CMP	Cytosine Monophosphate
CMT	Critical Micellization Temperature
CMV	Cytomegalovirus
CN	Coordination Number
cNRG	Cyclic Asparagine-Glycine-Arginine
CNT	Carbon Nanotube
cod	1,5-cyclooctadiene
Col	Collagen
Col$_h$	Columnar Hexagonal
Col$_r$	Columnar Rectangular
COM	Center of Mass
CoMoCat	Cobalt Molybdenum Catalyzed
Con A	Concanavalin A
CONTIN	Continuous Distributions of Exponen-tials
COR	Coronene
CORE	Component Resolved
CP	Cross-Polarization
Cp	Cyclopentadienyl
μCP	Microcontact Printing
m-CPBA	*meta*-Chloro-Perbenzoic Acid
CP-MAS	Cross-Polarized Magic-Angle Spinning
CPD	Cyclophanediene
CPK	Corey–Pauling–Koltun
CPL	Circularly Polarized Luminescence