

Supramolecular Chemistry

From Molecules to Nanomaterials

Concepts

Techniques

Molecular Recognition

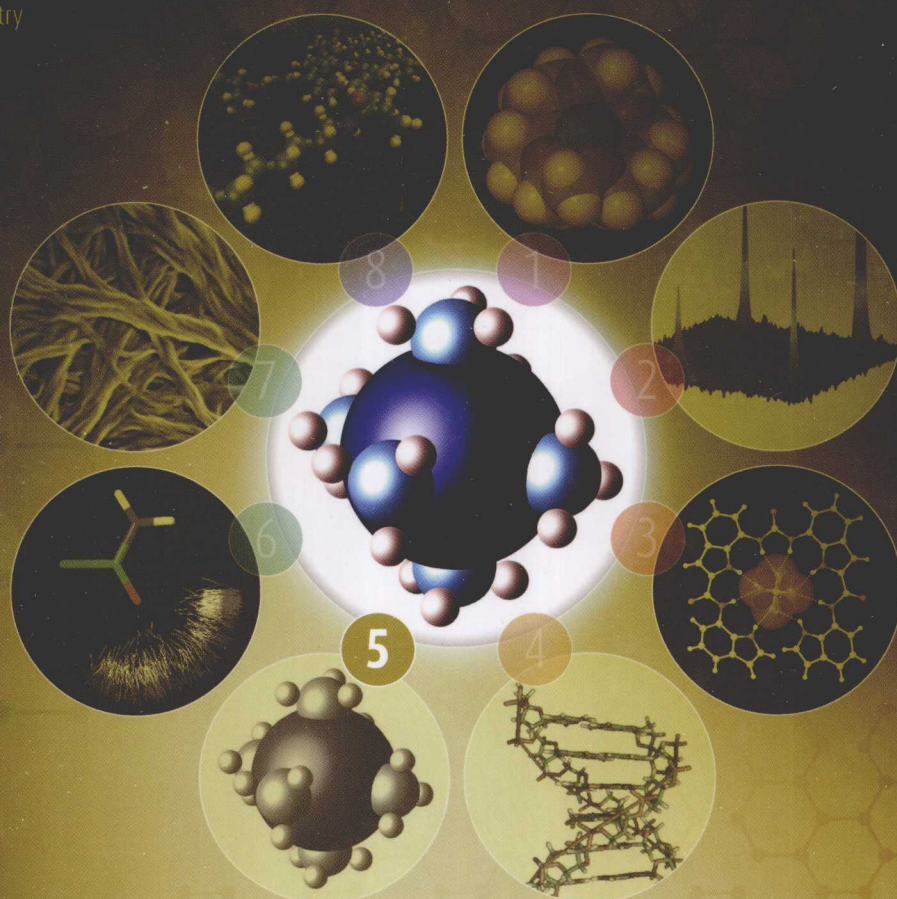
Supramolecular Catalysis, Reactivity and Chemical Biology

Self-Assembly and Supramolecular Devices

Supramolecular Materials Chemistry

Soft Matter

Nanotechnology



Editors Philip A. Gale and Jonathan W. Steed

Supramolecular Chemistry: From Molecules to Nanomaterials

Volume 5: Self-Assembly and Supramolecular Devices

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This edition first published 2012
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John Wiley & Sons Ltd, The Atrium, Southern Gate, Chichester, West Sussex, PO19 8SQ, United Kingdom

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Library of Congress Cataloging-in-Publication Data

Steed, Jonathan W., 1969-

Supramolecular chemistry : from molecules to nanomaterials / Jonathan W. Steed, Philip A. Gale. – 1st ed.
p. cm.

Includes bibliographical references and index.

ISBN 978-0-470-74640-0 (hardback)

1. Supramolecular chemistry. I. Gale, Philip A. II. Title.

QD878.S75 2012

547'.1226–dc23

2011039272

British Library Cataloguing in Publication Data

A catalogue record for this book is available from the British Library

ISBN: 978-0-470-74640-0

Typeset in 10/12.5 pt Times by Laserwords (Private) Limited, Chennai, India.
Printed and bound in Singapore by Markono Print Media Pte Ltd.

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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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March 2011

Abbreviations and Acronyms

17-AAG	17-Allylamino-17-demethoxygel-danamycin	ACE	Affinity Capillary Electrophoresis or Angiotensin Converting Enzyme
2-AP	2-Aminopyrimidine	ACh	Acetylcholine
9-ap	Anthracene-9-propionic Acid	ACHC	Aminocyclohexanecarboxylic Acid
4-bn-res	4-Benzylresorcinol	AChE	Acetylcholine Esterase
4,4'-bpe	<i>Trans</i> -1,2-bis(4-Pyridyl)Ethylene	AcOH	Acetic Acid
bpea	1,4-bis(4-Pyridyl)Ethane	ACR	Aza-Crown Resorcinarene
1,4-bpeb	1,4-bis[2-(4-Pyridyl)Ethenyl]benzene	aCTG	Triamino Cyclotriguaniacylene
bpee	1,4-bis(4-Pyridyl)Ethene	ACU	Undecyl-Aza-18-crown-6
1,4-bpef	<i>p</i> -Di-[2-(4-Pyridyl)Ethenyl]-2-fluorobenzene	AD	Acceptor Dendrimers or Activating Domain
bpp	1,3-bis(4-Pyridyl)Propane	Ad	Adamantane
1,5-bppo	Bis(4-Pyridyl)-1,4-pentadiene-3-one	Ad-PEG	Ad-Modified Polyethylene Glycol
2-CH ₃ THF	2-Methyltetrahydrofuran	ADA	Acceptor Donor Acceptor
4-Cl dpcb	<i>Rctt</i> -1,2-bis(4-Pyridyl)-3,4-bis(<i>p</i> -Chlorophenyl)Cyclobutane	ADMET	Acyclic Diene Metathesis
4-Cl stilbz	4-Chlorostilbazole	ADP	Adenosine 5'-Diphosphate
1D	One-Dimensional	ADR	Adriamycin
2D	Two-Dimensional	ADV	Adenovirus
3D	Three-Dimensional	aeg	<i>N</i> -(2-Aminoethyl)-Glycine
9EA	9-Ethyladenine	AEM	Arylene Ethynylene Macrocycle
6HB	Six-Helix Bundle	AFM	Atomic Force Microscopy
8HB	Eight-Helix Bundle	AFP	Alpha-Fetoprotein
2,3-nap	2,3-bis(4-Methylenethiopyridyl)-Naphthalene	AgNP	Silver Nanoparticle
1,8-nda	1,8-naphthalenedicarboxylic Acid	AHX	ϵ -aminohexanoic Acid
4-pa	(<i>E</i>)-3-(4-Pyridyl)Acrylic Acid	AIBN	2,2'-Azobis(Isobutyronitrile)
6PE	Sixfold Phenyl Embraces	AIEE	Aggregation-Induced Enhanced Emission
6PGL	6-Phosphogluconoylation	AK	Attenuated <i>K</i>
4-py-but	<i>Trans</i> -1,4-(4-Pyridyl)-1,3-butadiene	ALD	Atomic Layer Deposition
4-py-hex	<i>Trans</i> -1,6-(4-Pyridyl)-1,3,5-hexatriene	ALK	Ala-Leu-Lys-Arg-Gln-Gly-Arg-Thr-Leu-Tyr-Gly-Phe
4-vp	4-Vinylpyridine	ALP	Alkaline Phosphatase
AAO	Anodized Aluminum Oxide	ALP	Amphiphilic Lipopeptide
ABC	Adenosine-5'-triphosphate Binding Cassette	AM1	Austin Model 1
ABZ	Albendazole	AM1.5	Air Mass 1.5
AC	Alternating Current	AMF	Alternating Magnetic Field
ACA	Acetoxychavicol Acetate	AMFE	Anomalous Mole Fraction Effect
acac	Acetylacetonate	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	BET	Brunauer Emmett Teller or Back Electron Transfer
AO	Atomic Orbital	BFDMA	Bis-(11-Ferrocenylundecyl)-Dimethylammonium Bromide
AP	Aptamer-Photosensitizer	bFGF	Basic Fibroblast Growth Factor
APC	2,4-bis(4-Dialkylaminophenyl)-3-Hydroxy-4-alkylsulfanylcyclobut-2-enone	BH	Bcl-2 Homology
APED	Alternating Polyelectrolyte Deposition	BI	Bovine Insulin
API	Active Pharmaceutical Ingredient	BIA	Biomolecular Interaction Analysis
aPP	Avian Pancreatic Polypeptide	BIC	5-(Benzyloxy)-Isophthalic Acid
APTES	Aminopropyltriethoxysilane	BINAM	2,2'-diamino-1,1'-binaphthalene
AQ	Antraquinone	BINAP	2,2'-bis(Diphenylphosphino)-1,1'-binaphthyl
AR	Aromatic Resin	bipy	Bipyridine
AR	Aviram-Ratner	BiTE	Bispecific T-Cell Engager Molecules
ASGPR	Asialoglycoprotein Receptor	bix	1,4-bis(Imidazol-1-ylmethyl)-Benzene
Atb	S-2-Amino-4-trifluorobutyric Acid	BLM	"Black" Lipid Membrane
ATCh	Acetylthiocholine	BM	Ball Milling
ATP	Adenosine 5'-Triphosphate	BMP	Bone Morphogenetic Protein
ATR-FTIRS	Attenuated Total Reflection-Fourier Transform Infrared Spectroscopy	BNCT	Boron Neutron Capture Therapy
ATR-IR	Attenuated Total Reflectance Infrared	BNP	Binaphthyl Phosphate
ATRP	Atom Transfer Radical Polymerization	BNPP	Bis(4-Nitrophenyl)Phosphate
AU	Analytical Ultracentrifugation	BO	Butylene Oxide
Au-SNP	Au Supramolecular Nanoparticle	BODIPY	Boron Dipyrromethane
AUC	Analytical Ultracentrifugation	BoNT	<i>Botulinum Neurotoxin</i>
AuNP	Gold Nanoparticle	BP	Biphenol
AV	Ala-Val	BPA	Bipyridine Amine
AZTDP	Azido-3'-deoxythymidene 5'-diphosphate	BPB	Bromophenol Blue
AZTMP	3'-azido-3'-deoxythymidene 5'-monophosphate	BPEA	Bis-(Phenylethynyl)Anthracene
BAM	Brewster Angle Microscopy	bPP	Bovine Pancreatic Polypeptide
BAMP-ligand	Bis(aminomethyl)pyridine Ligand	BPP34C10	Bis(<i>p</i> -Phenylene)-34-Crown-10
BAPTA	1,2-bis(o-aminophenoxy)ethane- <i>N,N,N',N'</i> -tetraacetic acid	bpp-34-crown-10	Bisparaphenylene-34-crown-10
BAR	Barbituric Acid	BPPM	<i>N</i> -tert-butoxycarbonyl-4-diphenylphosphino-2-diphenylphosphino-methyl-pyrrolidine
BASE	Boron Affinity Saccharide Electrophoresis	bpy	Bipyridine
BASF	Baden Aniline and Soda Factory	BRGD-PA	Aspartate-Arg-Gly-Asp
BBV	Boronic Acid-Substituted Benzylviologen	BSA	Bovine Serum Albumin
BCA	Bio-bar-code amplification	BSM	Bovine Submaxillary Mucin
BCB	Benzocyclobutane	BSP	Bone Sialoprotein
BCC	Body Centered Cubic	BTA	Benzene-1,3,5-tricarboxamide
BCD	β -Cyclodextrin	BTB	1,3,5-benzenetribenzoate
BCP	Block Copolymer	BTC	1,3,5-benzenetricarboxylic Acid
bdc	1,4-benzenedicarboxylate	BTC	Benzene-1,3,5-tricarboxylate
BDC	Benzenedicarboxylic Acid	BTE	Backbone Thioester Exchange
BDE	Bond-Dissociation Energies	BTF6	1,2-bis(2-Methylbenzo[b]thiophen-3-yl)Hexafluorocyclopentene
BDG	Benzodiguanamine	BTM	Benzotetramisole
bdt	1,2,4,5-benzenetetracarboxylic Acid	BTMA	<i>n</i> -Butyltrimethylammonium
		BTX	Bent Triple-Crossover
		BZ	Belousov-Zhabotinsky
		BZD	Benzidine

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