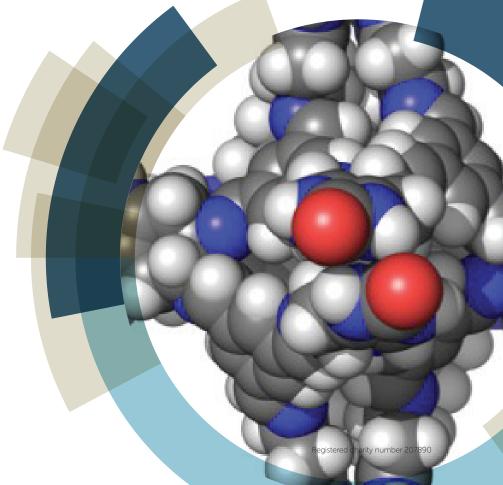




1 July 2014, Dublin, Ireland

# 5th Joint CSJ-RSC Symposium on Supramolecular Chemistry

# Programme Book



### Dear Colleagues

Welcome to the fifth in our series of Joint Chemical Society of Japan (CSJ) and Royal Society of Chemistry (RSC) symposia. This symposium is one of the activities jointly organized following the signing in 2010 of a five year International Cooperation Agreement between the CSJ and the RSC and we hope it will be another successful and memorable CSJ-RSC activity that will facilitate and foster collaborations and friendships. We are delighted to have this opportunity to organize this meeting in partnership with the CSJ and thank Mr Nobuyuki Kawashima, CSJ Executive Director and Professor Takashi Uemura from Kyoto University for all their help and support to make this event possible.

The Royal Society of Chemistry is the world's leading chemistry community, advancing excellence in the chemical sciences. With over 49,000 members and a knowledge business that spans the globe, we are the UK's professional body for chemical scientists, supporting and representing our members and bringing together chemical scientists from all over the world.

A not-for-profit organization with a heritage that spans 170 years, we have an ambitious international vision for the future. Around the world, we invest in educating future generations of scientists. We raise and maintain standards. We partner with industry and academia, promoting collaboration and innovation. We advise government on policy. And we promote the talent, information and ideas that lead to great advances in science.

In a complex and changing world, chemistry and the chemical sciences are essential. They are vital in our everyday lives and will be vital in helping the world respond to some of its biggest challenges. We're working to shape the future of the chemical sciences – for the benefit of science and humanity.

The RSC is also pleased to have the opportunity to work more closely with the Japanese chemical science community through Dr Hiromitsu Urakami, our Representative for Japan, who is based full-time in our office in Tokyo. This office is located in the CSJ building in Tokyo and we are very grateful to the CSJ for all their help and advice in establishing this office.

Each of the speakers in this symposium is an international expert in the area of supramolecular chemistry. We hope that the presentations will stimulate the exchange of ideas and experiences between all participants. We thank each of the speakers and all the participants for their contributions to this symposium.

Once again a very warm welcome to what promises to be an exciting scientific event. We hope that the symposium will provide a springboard for future activities and that it will foster new research collaborations. We look forward to continuing our close partnership with the CSJ with more joint CSJ-RSC activities in the future.



Professor Lesley Yellowlees CBE President Royal Society of Chemistry Dear Colleagues,

In 2007, the Chemical Society of Japan (CSJ) began holding joint symposia with the Royal Society of Chemistry (RSC) as a part of its efforts to promote international exchange. The symposia were held alternatingly in Japan and in the United Kingdom in 2007, 2008, 2010, and 2012. Now CSJ and its 30,000 members are delighted to hold the fifth joint symposium in Dublin, Ireland, in conjunction with the 13th International Symposium on Advancing the Chemical Sciences (ISACS13).

The world is now confronting global-scale challenges in environment, resource, energy, food, and water, among others, resulting from the population growth and human activities. Given that it is practically impossible for any single nation to find solutions for these issues, all countries must cooperate in creating innovations based on science and technology. As CSJ President, I believe that chemistry is a key to such innovations where chemists can play significant roles. As one of the main action plans for 2014, therefore, CSJ is enhancing collaboration with the chemistry-related societies around the world.

CSJ and RSC have already begun increasingly close collaboration in recent years as reflected in our International Cooperation Agreement established in 2010. More specifically, for example, the two societies started working together to improve chemistry-related capabilities in both countries by exchanging research findings at the two symposia in London, with leading researchers presenting Japanese and European technological innovations in chemistry, particularly the areas of green and life-related innovations: The first symposium in March 2013 on lithium-ion batteries and the second in March 2014 on peptide drugs.

*Supramolecular chemistry*, the topic of the 2014 joint symposium, is a highly advanced interdisciplinary field that covers the chemical, physical and biological properties and functions of supramolecules, complex chemical materials formed through specific intermolecular interaction beyond individual molecules. Recently we have been witnessing rapid advances in various research areas related to supramolecular chemistry, thus leading to a diverse range of materials that exhibit electrical conductivity, luminescence, self-healing, catalysis, and separation functions, among many others. The development of applications of these materials is generating considerable excitement.

To this fifth symposium, six distinguished scientists in the area of supramolecular chemistry are invited to present their research results and to exchange information and ideas:

- Dr. Thomas Bennett, University of Cambridge
- Dr. Kim Jelfs, Imperial College London
- Dr. Rie Makiura, Osaka Prefecture University
- Dr. Wolfgang Schmitt, University of Dublin, Trinity College
- Dr Yoshinori Takashima, Osaka University
- Dr. Takashi Uemura, Kyoto University

I hope that this joint symposium will contribute to accelerating the research in supramolecular chemistry and related domains and also to strengthening the ties between the chemistry communities in the United Kingdom & Ireland, and Japan as represented by RSC and CSJ.



Dr. Sadayuki Sakakibara President The Chemical Society of Japan

### 5th Joint CSJ-RSC Symposium on Supramolecular Chemistry

### **Chartered Accountants House, Dublin**

### Tuesday 1st July, 2014

Time	Event	
13.30	Introduction and welcome Nobuyuki Kawashima, CEO, <i>Chemical Society of Japan, Japan</i> Robert Parker, CEO, <i>Royal Society of Chemistry, UK</i>	Chair Sarah Thomas Royal Society of Chemistry, UK
13:50	Stimuli responsive and self-healing supramolecular materials through host and guest interactions Yoshinori Takashima, <i>Osaka University, Japan</i>	<b>Chair</b> Takashi Uemura <i>Kyoto University,</i> Japan
14:20	Amorphous metal-organic frameworks: Structure, properties and applications Thomas D Bennett, University of Cambridge, UK	
14:50	Surface nanotectonics of metal-organic frameworks at air/liquid interfaces Rie Makiura, Osaka Prefecture University, Japan	
15:20	Coffee break	
15:50	Molecular prediction of porous organic molecules Kim E Jelfs, Imperial College London, UK	Chair Thomas D Bennett <i>University of</i> <i>Cambridge, UK</i>
16:20	Polymer chemistry in coordination nanochannels Takashi Uemura, Kyoto University, Japan	
16:50	Supramolecular approaches to nanoscopic coordination cages, clusters and high- surface area coordination networks Wolfgang Schmitt, <i>University of Dublin, Ireland</i>	
17.20	Closing remarks Lesley Yellowlees, President, Royal Society of Chemistry, UK	Chair Sarah Thomas, Royal Society of Chemistry, UK
	Close	

Notes



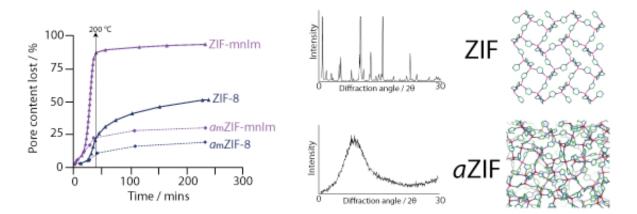
Thomas Douglas Bennett University of Cambridge, UK Tdb35@cam.ac.uk

Thomas D. Bennett was born in South Shields, UK (1986).He read the Natural Sciences Tripos at the University of Cambridge, specializing in Chemistry, where he undertook research on the synthesis of 'light' metal-organic frameworks under Dr. Paul T. Wood.After moving to the Department of Materials Science and Metallurgy, he completed a PhD on the thermo-mechanical Properties of Zeolitic Imidazolate Frameworks, under Professor Anthony Cheetham. In 2009 he was awarded an honorary research fellowship at the Ras Al Khaimah center for advanced materials (RAK-CAM), and in 2012 he won the Panalytical prize for an outstanding contribution to X-ray diffraction.Thomas was elected as a Research Fellow at Trinity Hall, University of Cambridge in 2013.He currently focuses on the synthesis, structure, properties and applications of amorphous hybrid frameworks, alongside the formation of hybrid glasses.

# Amorphous metal-organic frameworks: Structure, properties and applications

### Thomas Douglas Bennett University of Cambridge, UK

A small but growing number of cases of MOFs lacking the long-range order characteristic of crystalline structures are steadily capturing scientific interest (though they remain woefully under represented), despite the phenomenon being known in inorganic zeolites. The majority of work has been performed on a sub-family of MOFs called zeolitic imidazolate frameworks (ZIFs), which consist of tetrahedral metal centers (typically  $Zn^{2+}$  or  $Co^{2+}$ ), connected by Imidazolate-type ( $C_3H_3N_2^{-1}$ ) organic ligands in zeolitic topologies. Recently, we discovered that ZIFs could be amorphized by ball-milling and heating, and that the resultant amorphous material has a similar topology to that of silica glass.<sup>1</sup> Similarly, they can also be amorphized by pressure, though there is considerable debate about the reversibility of the process.



Amorphous MOFs, whilst significantly harder to characterise, are more mechanically stable than their well-known crystalline counterparts. They also offer routes to functional glass materials, through melting of functionalised parent frameworks (incoporating lanthanide ions or chiral ligands). Some of these glasses have been shown to be topologically 'perfect' with fragilities lower than that of aSiO<sub>2</sub>. Recently, we have shown that the release *in-vitro* of ibuprofen and caffeine stored within ZIF-8 can be tailored by ball-milling inducing collapse of the framework, whilst I<sub>2</sub> has also been irreversible captured using similar methodology.<sup>2</sup>

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Kim Jelfs Imperial College London, UK k.jelfs@imperial.ac.uk

Since October 2013 I have held a Royal Society University Research Fellowship (URF) at Imperial College London, where I am now establishing my own independent research group. My fellowship is entitled "Directing the synthesis of functional molecular materials". I completed my PhD in Computational Chemistry at UCL, working on the development and application of modelling to understand zeolite crystal growth and was awarded the Ramsay Medal for the best completing PhD student. I subsequently became a visiting researcher at the Universitat de Barcelona for 6 months, working with Prof. Stefan Bromley on the prediction of silicate cluster formation, before moving to the University of Liverpool, working as a PDRA across the experimental groups of Profs. Matt Rosseinsky and Andy Cooper. I focused upon modelling porous materials, with my expertise spanning zeolites, metal-organic frameworks, polymers and porous molecular materials. Research highlights over my career thus far include high impact publications in Nature, Nature Chemistry, Angewandte Chemie and JACS. In several of my publications to date I have been the sole computational author, providing both insight and predictions for the experimental research.

### Molecular prediction of porous organic molecules

Kim E. Jelfs<sup>a\*</sup>, Ming Liu<sup>b</sup> and Andrew I. Cooper<sup>b</sup> <sup>a</sup>Department of Chemistry, Imperial College London, UK <sup>b</sup>Department of Chemistry, University of Liverpool, UK

The modular assembly of discrete, intrinsically porous organic molecules is an alternative to forming porous materials through extended networks that are chemically bonded in 3-dimensions.<sup>1</sup> These modular materials can have advantages, such as being easily processable,<sup>2</sup> co-crystallisation allowing property tuning<sup>3</sup> and their greater mobility facilitating stimuli-response behaviour, such as "on"/"off" porosity switching.<sup>4</sup> From a computational perspective, it has been shown that crystal structure prediction techniques can rationalise the observed polymorphs<sup>3</sup> and we have used molecular dynamics simulations to explain observed sorbate selectivity.<sup>4,5</sup> More recently, we have been developing an approach towards in silico design of new porous molecular materials with desired structures and properties. This automates the assembly and screening of hypothetical molecules from a library of precursors. The first goal is to predict the conformation of the molecule that will be formed. It is crucial to test whether the molecule will be "shape persistent", keeping an internal void in the absence of solvent, as we have previously spent months synthesising large molecules that collapse upon desolvation. The second goal is to correctly predict which molecule will form from a pair of precursors - e.g. will a [2+3], [4+6], [6+9] or [8+12] reaction occur? We have demonstrated the successful prediction of an odd-even effect for a series of alkane diamines (a).<sup>6</sup> Further, we can predict how post-synthetic modification can engineer shape-persistence and acid and base-resistance (b), as well as using computational predictions to engineer novel shaped molecules and to perform rapid computational screening to predict properties for hypothetical molecules (c).

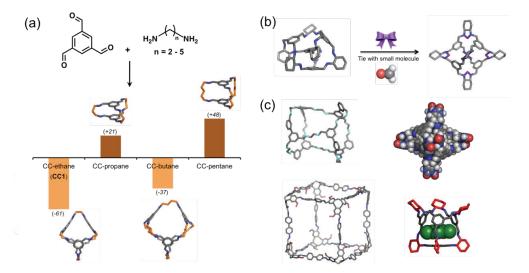


Figure: a) Size and structure prediction of an odd-even alkane chain effect; b) Predictions of post-synthetic modification routes to engineer shape persistence; c) Predictions of promising synthetic targets for new topologies and functions.

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**Rie Makiura** *Osaka Prefecture University, Japan* r-makiura@21c.osakafu-u.ac.jp

Rie Makiura graduated from the University of Tsukuba in 2002 (MSc, Chemistry). Following an extended spell as a Researcher at the Frontier Device Research Center of SEIKO EPSON Cooperation in Nagano (2002 - 2007) where she worked on the development of organic electronic (OLED, TFT) devices and thin film all-solid batteries, she returned to academia as an Assistant Professor at the Department of Chemistry, Kyushu University (2007 - 2010). She was awarded her D.Sc. degree from Kyushu University in 2010. In April 2010, she was appointed as a tenure-track Lecturer at the Nanoscience and Nanotechnology Research Center, Osaka Prefecture University. She was also appointed as a PRESTO (Precursory Research for Embryonic Science and Technology) researcher, the Japan Science and Technology (JST) agency in 2012. She received the 2013 Award for Outstanding Young Women Chemists by the Chemical Society of Japan and the 2014 Young Scientists' Prize of the Commendation for Science and Technology by the Minister of Education, Culture, Sports, Science and Technology, Japan. Her research focus is currently on the rational design and construction of functional surface/interface nanostructures.

# Surface nanotectonics of metal-organic frameworks at air/liquid interfaces

Rie Makiura<sup>1,2</sup>

<sup>1</sup> Nanoscience and Nanotechnology Research Center, Osaka Prefecture University, 1-2 Gakuen-cho, Naka-ku, Sakai, Osaka 599-8570 JAPAN <sup>2</sup> JST-PRESTO, Email: r-makiura@21c.osakafu-u.ac.jp HP: http://www.nanosq.21c.osakafu-u.ac.jp/ttsl\_lab/r\_makiura/index.html

Development of rational methods for creating ordered structures with nanometer scale precision is one of the central issues in the nanoscience and nanotechnology fields. Molecular based nanoassemblies are of great interest because of their rich variety of structures and facile functionalization. We have succeeded to assemble perfect preferentially-oriented metal-organic framework (MOF) nanofilms by developing a facile bottom-up fabrication at liquid-air interfaces <sup>1-3</sup>. Employing liquid surfaces in order to tune 2D growth is highly promising as spontaneous organizational features of the building components can be enhanced. In order to develop further the strategy to grow 2D molecular assemblies with fine control of both the domain size and the molecular arrangement, understanding the formation mechanism associated with the coordinative linkages is necessary.

Here I will present a discussion of the formation process of a 2D MOF array (NAFS-13) on a liquid surface by varying the fabrication conditions with the structural details obtained by the grazing incidence synchrotron X-ray diffraction (GIXRD) technique at the air/water interface (Fig. 1). A porphyrin derivative (MTCPP) was selected as the molecular building unit. The results reveal that the highly-crystalline preferentially-oriented ultra-narrow NAFS-13 sheets form immediately after spreading the molecular building units on the copper ion aqueous solution subphase (Fig. 1). Following the insight obtained on the nanosheet formation process, a new approach of sheet assembly – post-injection of the metal linkers into the subphase – was developed that effectively led to the enlargement of the NAFS-13 sheet domain size <sup>4-5</sup>.

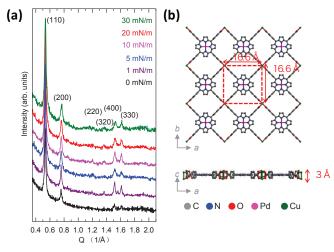


Fig. 1 Structural evolution of the MTCPP-Cu nanosheets on liquid surfaces. (a) GIXRD ( $\lambda$ =1.549 Å) profiles with change in surface pressure. (b) Schematic diagram of the crystalline structure of NAFS-13.

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Wolfgang Schmitt School of Chemistry, Trinity College Dublin, Ireland schmittw@tcd.ie

Wolfgang Schmitt is currently an Associate Professor and working in the area of Inorganic and Materials Chemistry at Trinity College Dublin. He studied Chemistry at the Technical University Darmstadt (TU), Germany and graduated as Dipl. Ing. (Chemistry) in 1999. He was awarded the Dr. Anton Keller Prize from the University of Darmstadt and received his Ph.D. from the University of Karlsruhe in 2002. From 2002-2003 he worked as a postdoctoral fellow at the Royal Institution of Great Britain. In March 2004 he was selected to join the International Centre for Young Scientists at the National Institute for Materials Sciences in Tsukuba, Japan as an independent research fellow. After joining Trinity College Dublin he was elected as 'Fellow' in 2009. He is an SFI Principal Investigator and received further funding from SFI, EPA and IRCSET. Dr. Schmitt's research interests focus on the supramolecular chemistry of hybrid organic-inorganic materials aiming to exploit these advanced materials for sustainable energy applications, catalysis and molecular magnetic devices. Current activities focus on metal-organic frameworks (MOFs) for hydrogen storage applications, CO<sub>2</sub> fixation and activation, bio-mimetic syntheses, self-assembly processes, synthetic approaches to nanoscopic clusters (polyoxometalates and other coordination clusters) and molecular coordination cages.

# Supramolecular approaches to nanoscopic coordination cages, clusters and high-surface area coordination networks

Wolfgang Schmitt<sup>\*</sup>, Lei Zhang, Nianyong Zhu and Camelia I. Onet University of Dublin, Trinity College Dublin, School of Chemistry, Dublin, D2, Ireland

Coordination clusters and molecular capsules are supramolecular compounds that have received considerable attention over the last years. The arrangement and connectivity of the metal centres in such nanoscopic assemblies may result in unusual magnetic and electronic attributes whist the resulting inner cavities provide unique, restricted chemical environments which allow the molecules to be applied for molecular recognition purposes, as supramolecular containers or as catalysts.<sup>1</sup> We studied the formation of V<sup>V</sup>/V<sup>IV</sup> cages which form upon partial reduction of vanadates in the presence of organic ligands and identified new tetra- and pentanuclear building units and utilized these pre-organized structural motifs to produce novel hybrid capsules,  $[H_2V_{10}O_{18}(L)_4]^{8-}$  with controllable dimensions (Fig.1, *left*).<sup>2</sup> Our investigations further demonstrate that template aggregates can be utilised to direct the assembly process of the tetranuclear building unitsallowing a successive built-up of highly symmetrical nanosopic coordination cages that contain between 20 and 32 V and As centres.<sup>3</sup> Our research highlights the relation between Platonic Templates and Archimedean cage structures (Fig.1, *right*). We demonstrated that the synthetic approach can be applied to prepare nanoscopic Mn coordination clusters that give rise to interesting molecular magnetic properties.<sup>4</sup> Other aspects focus on self-assembly processes that yield carboxylate-based MOFs that reveal high surface areas and can be used for gas storage applications.<sup>5</sup>

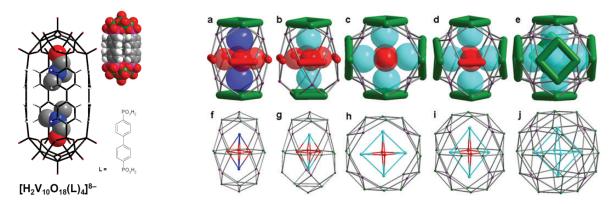


Fig 1. Left: Hybrid vanadate capsules; Right: Simplified core structures of vanadate coordination cages and their encapsulated templates.

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Yoshinori Takashima was born in 1974 in Osaka, Japan.He received his Ph.D. degree in 2002 from Osaka University, under the guidance of Prof. Akira Harada (Osaka Univ.), focusing on the polymerization catalyst using group 4 transition metal catalyst.He was selected as a Research Fellow of the Japan Society for the Promotion of Science (JSPS) in 1999–2002.During the winter of 2003, as a visiting fellow, he worked at Prof. Jeffrey M. Stryker's lab, Department of Chemistry, the Univ. of Alberta.In 2003, he obtained his PhD in Polymer Science from Osaka Univ.Prior to joining the Graduate School of Science at Osaka Univ. in 2004 as an assistant professor for Professor Akira Harada, he served as a postdoctoral research fellow from JSPS in the Division of Applied Chemistry, Osaka Univ. for Professor Toshikazu Hirao. In 2011, he worked at Prof. Philip A. Gale's lab, Department of Chemistry, the Univ. of Southampton as a visiting researcher.His research interests include supramolecular materials, catalysts, molecular motors and devices.

## Stimuli responsive and self-healing supramolecular materials through host and guest interactions

### Yoshinori Takashima

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In biological system, there are some attractive functions such as enzymatic reactions, muscle motion, and self-healing properties. Inspired by these functions, many supramolecular chemists engage the creation of supramolecular complexes reminescent of biological functions. We have focused on the creation of supramolecular materials reminescence of the muscle motion, self-organization of cell and self-healing properties through host-guest interactions (Fig. 1). We employ cyclodextrin (CD) as a host molecule because it is environmentally benign and has diverse applications. This presentation demonstrates as following topics.

#### Supramolecular actuator

To prepare supramolecular actuators, azobenzene and ferrocene derivatives were chosen as photo and redox-responsive guest molecules. The chemical crosslinked hydrogel with stimuli-responsive groups showed the bending behavior by external stimuli, which controlled the ratio of inclusion complexes as a function of crosslinkers.<sup>1-3</sup>

#### Stimuli-responsive self-healing materials

We have investigated two approaches to prepare self-healing materials. First method is a supramolecular materials using poly(acrylic acid) modified with cyclodextrins (pAA-CDs) as a host polymer and pAA with ferrocene (pAA-Fc) as a guest polymer (Fig. 2).<sup>4</sup> Second method is the radical copolymerization of  $\beta$ CD and adamantane (Ad) monomers to give supramolecular hydrogels in water without crosslinkers (Fig. 3). The supramolecular hydrogels formed by host-guest interactions exhibits self-healing properties to reach 99% of the initial gel's strength.<sup>5</sup>

#### Macroscopic self-assembly through non-covalent bond

We achieved macroscopic self-assemblies through molecular recognition of CDs, which can be used to direct the assembly of macroscopic objects into larger aggregated structures.<sup>6</sup> Here we report an A-B-C type redox–responsive gel assembly system based on host–guest interactions between CD and Fc / cation–anion interactions between Fc<sup>+</sup> and sodium *p*–styrenesulfonate (SSNa).<sup>7-8</sup>Such macroscopic assemblies remind us of the selective formation of cell organization.

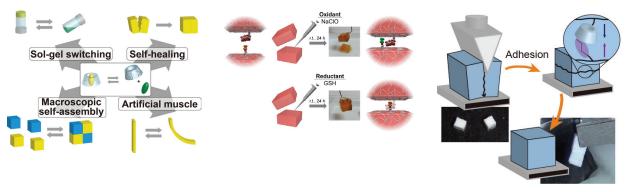


Figure 1. Conceptual scheme of our studies.

Figure 2. Redox-responsive healing experiment of the pAA-6βCD/pAA-Fc hydrogel using oxidizing/reducing agents.

Figure 3.Self-healing property of the βCD-Ad gel.

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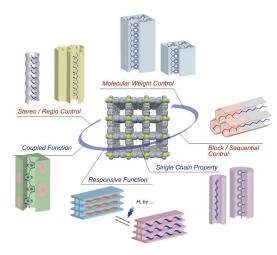
Takashi Uemura received his PhD under the supervision of Prof. Yoshiki Chujo at Kyoto University in 2002. During that time, he joined Prof. Christopher K. Ober group at Cornell University as a summer student (1999). He became an assistant professor in Prof. Susumu Kitagawa group at Kyoto University (2002), and was promoted associate professor (2010). He has also worked as a researcher in PRESTO program during 2006-2010 and was recently selected as a research director in CREST program of Japan Science and Technology Agency (JST). He received many awards including the Chemical Society of Japan Award for Young Chemists (2010) and the Commendation for Science and Technology by the Minister of Education, Culture, Sports, and Technology, the Young Scientists' Prize (2013). His research interest focuses on the preparation and property of synergistic nanohybrids between coordination compounds and polymer materials, in particular, polymer chemistry in confined coordination frameworks.

### Polymer chemistry in coordination nanochannels

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Recently, Metal-Organic Frameworks (MOFs) composed of metal ions and organic ligands have been extensively studied. The characteristic features of MOFs are highly regular channel structures, controllable channel sizes approximating molecular dimensions, and designable surface functionality. Use of their regulated and tunable channels for a field of polymerization can allow multi-level controls of the resulting polymer structures (Figure).<sup>1</sup> In addition, construction of nanocomposites between MOFs and polymers will provide unprecedented material platforms to accomplish many nanoscale functions (Figure).<sup>1</sup>



Controlled radical polymerization of vinyl monomers can be attained in MOF nanochannels.<sup>2</sup> Considerable effects of pore size, shape, and functionality on the resulting polymer structure and assembly were demonstrated. A variety of polymerization reactions can be also catalyzed in MOFs, providing functional poly(substituted acetylene)s, polypyrrole, and so on.<sup>3</sup> We also studied the properties of polymer chains confined in the nanochannels of MOFs, which led to produce many functional nanocomposite materials including sensor, adsorbent, and conductor.<sup>4</sup> New application of a MOF-peptide composite for an autonomous motor was also performed.<sup>5</sup>

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Notes





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