



The following Communications have been judged by at least two referees to be “very important papers” and will be published online at www.angewandte.org soon:

D. V. Esposito, S. T. Hunt, A. L. Stottlmyer, K. D. Dobson,
B. E. McCandless, R. W. Birkmire, J. G. Chen*

**Low-Cost Hydrogen-Evolution Catalysts Based on Monolayer
Platinum on Tungsten Monocarbide (WC) Substrates**

R. Matsui, K. Seto, K. Fujita, T. Suzuki, A. Nakazaki, S. Kobayashi*
**Unusually E-Selective Ring-Closing Metathesis to Form
Eight-Membered Rings**

J. S. Chen, T. Zhu, C. M. Li, X. W. Lou*

Building Hematite Nanostructures Using Oriented Attachment

S. Sun, G. Zhang, D. Geng, Y. Chen, R. Li, M. Cai, X. Sun*
**A New Highly Durable Platinum Nanocatalyst for PEM Fuel
Cells: Multiarmed Star-like Nanowire Single Crystals**

Y. Matsuki, M. T. Eddy, R. G. Griffin, J. Herzfeld*
Rapid 3D MAS NMR Spectroscopy at Critical Sensitivity

T. J. Hebden, A. J. S. John, D. G. Gusev, W. Kaminsky, K. I. Goldberg,
D. M. Heinekey*

Preparation of a Dihydrogen Complex of Cobalt

Y. Zhang, G. M. Miyake, E. Y.-X. Chen*

**Alane-Based Classical and Frustrated Lewis Pairs in Polymer
Synthesis: Rapid Polymerization of Methyl Methacrylate and
Naturally Renewable Methylene Butyrolactones to
High-Molecular-Weight Polymers**

K. Breuker,* S. Brüscheweiler, M. Tollinger

**Electrostatic Stabilization of Native Protein Structure in the Gas
Phase**

J. Zeng, X. Xia, M. Rycenga, P. Henneghan, Q. Li, Y. Xia*
**Successive Deposition of Silver on Silver Nanoplates: Lateral
Versus Vertical Growth**



“When I was eighteen I wanted to be an engineer.
When I wake up I wake up my son and my daughter...”
This and more about Laurent Maron can be found on
page 9562.

Author Profile

Laurent Maron _____ 9562

Bert L. Vallee 1919–2010

Obituaries

W. Maret _____ 9563 – 9564

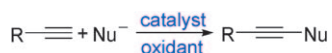
Metal–Organic Frameworks

Leonard R. MacGillivray

Books

reviewed by S. Kaskel _____ 9565

Oxidative heterocoupling of terminal alkynes mediated by metal complexes has emerged as a promising new strategy for the incorporation of alkynyl functionality into organic molecules (see scheme). Recent key developments in the construction of $C_{sp}-C_{sp^3}$, $C_{sp}-C_{sp^2}$, $C_{sp}-C_{sp}$, and C_{sp} –heteroatom bonds are highlighted.



Highlights

Cross-Coupling

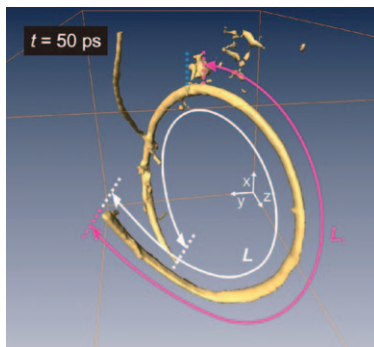
Z. Shao,* F. Peng _____ 9566 – 9568

**Metal-Mediated Oxidative Cross-Coupling
of Terminal Alkynes: A Promising Strategy
for Alkyne Synthesis**

Electron Tomography

D. S. Su* _____ 9569–9571

Electron Tomography: From 3D Statics to 4D Dynamics



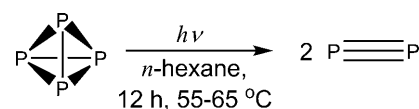
Time and motion studies: 4D electron tomography enables the direct investigation of transient states of materials, structural dynamics of large molecular objects (for example the motion of a spiral carbon nanotube), and biological systems under controlled conditions.

Diphosphorus

C. A. Russell _____ 9572–9573

P≡P, a Laboratory Reagent?

Two P or not two P? This bastardization of a well-known Shakespearean phrase is used to highlight recent work that shows that irradiation of white phosphorus produces diphosphorus, P₂ (see scheme), which may be reacted in situ in a double Diels–Alder reaction to give cage diphosphines.

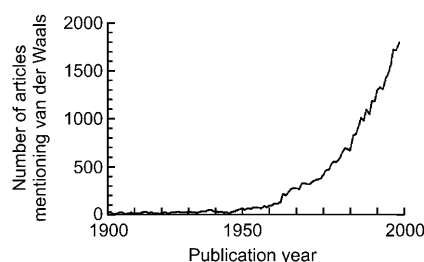


Essays

van der Waals

K. T. Tang,* J. P. Toennies* _ 9574–9579

Johannes Diderik van der Waals: A Pioneer in the Molecular Sciences and Nobel Prize Winner in 1910



Evergreen: The work of Johannes Diderik van der Waals continues to have an impact in the sciences even 100 years after he was awarded the Nobel Prize in Physics for “his work on the equation of state for gases and liquids”. His name is associated with many modern physical concepts in the molecular sciences, as the adjacent plot, referring to articles in which “van der Waals” is mentioned in the title, abstract, or keywords shows.

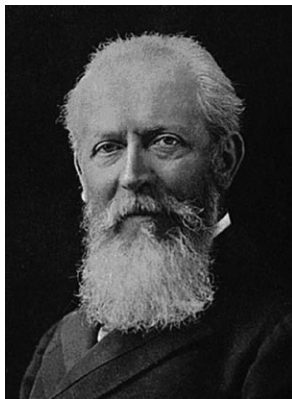
For the USA and Canada:

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individuals who are personal members of a national chemical society prices are available on request. Postage and handling charges included. All prices are subject to local VAT/sales tax.

A blast from the past: Through his contributions on derivatization and structural elucidation Otto Wallach revolutionized terpene chemistry. His research in this area, begun in 1884, earned him the Nobel Prize in Chemistry a hundred years ago.

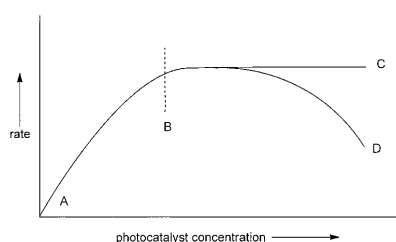


Terpenes

M. Christmann* _____ 9580–9586

Otto Wallach: Founder of Terpene Chemistry and Nobel Laureate 1910

Breaking the impasse: Earlier this year Maschmeyer and Che outlined an approach for normalizing and comparing the apparent quantum yields (AQY) for hydrogen photogeneration catalyzed by semiconductor powders. In the plot of photocatalyst concentration versus AQY they proposed that the linear region was the most appropriate region for comparison. In his Correspondence Kisch refutes this proposal and states that the area at the onset of the plateau region (B) in a plot of catalyst concentration versus reaction rate is relevant. Maschmeyer and Che explain their rationale and conclude that the discussion has arisen because of different emphases in studying photocatalysis.



Correspondence

Normalizing Photocatalysis

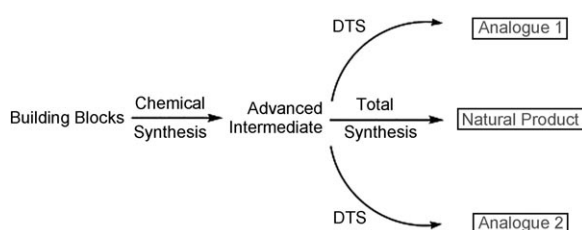
H. Kisch* _____ 9588–9589

On the Problem of Comparing Rates or Apparent Quantum Yields in Heterogeneous Photocatalysis

Normalizing Photocatalysis

T. Maschmeyer,* M. Che ____ 9590–9591

Intrinsic Catalytic Activity versus Effective Light Usage—A Reply to Professor Kisch's Comments



Editor's choice: Molecular editing of natural products through diverted total synthesis (DTS) is a powerful approach that offers far-reaching opportunities for discovery at the interface of biology and

chemistry. This Review assembles a collection of classic and new cases that illustrate and underscore the scientific possibilities for practitioners of chemical synthesis.

Reviews

Molecular Editing

A. M. Szpilman,*
E. M. Carreira* _____ 9592–9628

Probing the Biology of Natural Products: Molecular Editing by Diverted Total Synthesis

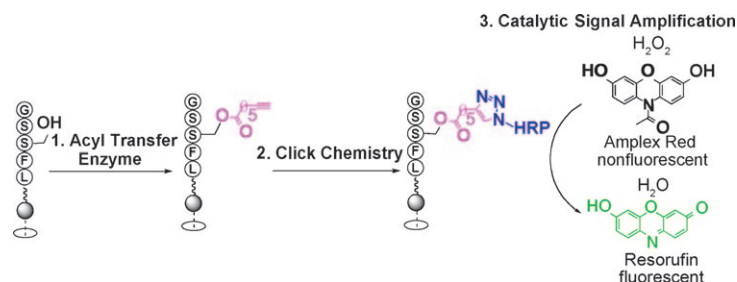
Communications

Enzyme Assays

A. L. Garner, K. D. Janda* — 9630–9634



cat-ELCCA: A Robust Method To Monitor the Fatty Acid Acyltransferase Activity of Ghrelin O-Acyltransferase (GOAT)



Assays armed with catalytic signal amplification have arisen as superior systems for ultrasensitive detection of analytes. A conceptually new enzyme assay called cat-ELCCA (catalytic assay using enzyme-linked click-chemistry) is described, in

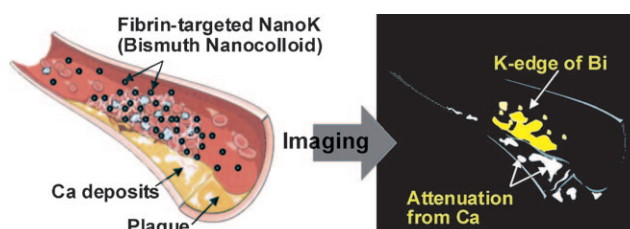
which an enzyme-linked azide is utilized to arm the assay with catalytic fluorescence signal amplification. By using this assay technology, the first high-throughput screen for recently disclosed ghrelin O-acyltransferase (GOAT) was developed.

Functional Nanocolloids

D. Pan,* E. Roessl, J.-P. Schlomka, S. D. Caruthers, A. Senpan, M. J. Scott, J. S. Allen, H. Zhang, G. Hu, P. J. Gaffney, E. T. Choi, V. Rasche, S. A. Wickline, R. Proksa, G. M. Lanza* — 9635–9639



Computed Tomography in Color: NanoK-Enhanced Spectral CT Molecular Imaging



Multicolored imaging: A new class of molecular imaging agent has been developed based on low-molecular-weight organically soluble bismuth to detect and

quantify intraluminal fibrin presented by ruptured plaque in the context of computed tomography angiograms without calcium interference.

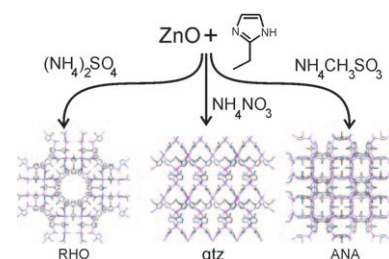
Mechanosynthesis

P. J. Beldon, L. Fábián, R. S. Stein, A. Thirumurugan, A. K. Cheetham, T. Friščić* — 9640–9643



Rapid Room-Temperature Synthesis of Zeolitic Imidazolate Frameworks by Using Mechanochemistry

Freshly ground: Improved mechanochemical methodologies, such as liquid-assisted grinding and ion- and liquid-assisted grinding enable the rapid and topologically selective synthesis of porous and nonporous zeolitic imidazolate frameworks with diverse topologies, at room temperature and directly from zinc oxide (see scheme).

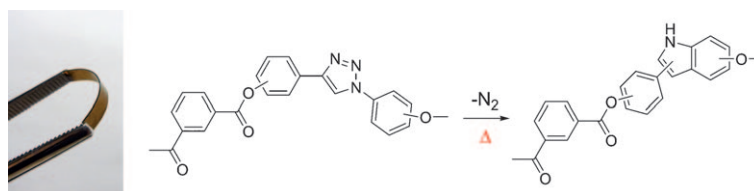


Smart Polymers

B.-Y. Ryu, T. Emrick* — 9644–9647



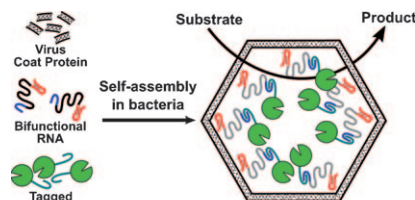
Thermally Induced Structural Transformation of Bisphenol-1,2,3-triazole Polymers: Smart, Self-Extinguishing Materials



Safer plastics: A novel class of polymers, termed bisphenol-1,2,3-triazole (BPT) polyarylates, was prepared by polycondensation chemistry. They exhibit high-

performance and self-extinguishing properties as a result of their heterocyclic nature and a thermally induced structural transformation (see picture).

Packaged molecular machines are available using dual expression vectors that guide the preparation of Q β virus-like particles encapsulating multiple enzymes. Packaging is promoted by RNA aptamer sequences that bridge between the coat protein and a peptide tag fused to the desired cargo (see scheme). Peptidase E and luciferase were thus encapsulated and shown to be catalytically active inside the particle.



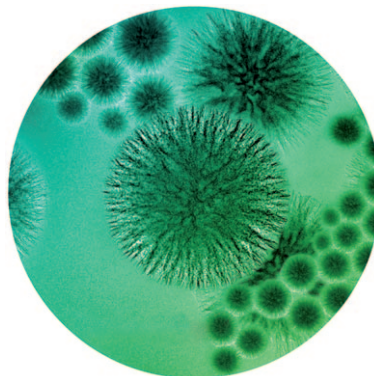
Catalytic Virus-like Particles

J. D. Fiedler, S. D. Brown, J. L. Lau,
M. G. Finn* 9648–9651

RNA-Directed Packaging of Enzymes
within Virus-like Particles



Fibrous nanosilica: A new family of high-surface-area silica nanospheres (KCC-1) have been prepared (see picture). KCC-1 features excellent physical properties, including high surface area, unprecedented fibrous surface morphology, high thermal (up to 950°C) and hydrothermal stabilities, and high mechanical stability.



Fibrous Nanomaterials

V. Polshettiwar,* D. Cha, X. Zhang,
J. M. Basset* 9652–9656

High-Surface-Area Silica Nanospheres
(KCC-1) with a Fibrous Morphology



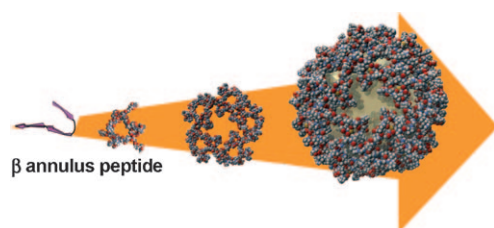
Shedding light: Nitroaromatic compounds on gold nanoparticles (3 wt%) supported on ZrO₂ can be reduced directly to the corresponding azo compounds when illuminated with visible light

or ultraviolet light at 40°C (see picture). The process occurs with high selectivity and at ambient temperature and pressure, and enables the selection of intermediates that are unstable in thermal reactions.

Photochemistry

H. Y. Zhu,* X. B. Ke, X. Z. Yang, S. Sarina,
H. W. Liu 9657–9661

Reduction of Nitroaromatic Compounds
on Supported Gold Nanoparticles by
Visible and Ultraviolet Light



Assemble a virus: A chemical strategy to de novo design “tailor-made” viruslike nanocapsules was developed (see picture). 24-mer β -annulus peptides of the tomato bushy stunt virus (TBSV) sponta-

neously self-assemble into hollow nanocapsules with a size of 30–50 nm. The hollow structure of the assemblies was clearly revealed by small-angle X-ray scattering (SAXS).

Viral Capsids

K. Matsuura,* K. Watanabe, T. Matsuzaki,
K. Sakurai, N. Kimizuka 9662–9665

Self-Assembled Synthetic Viral Capsids
from a 24-mer Viral Peptide Fragment



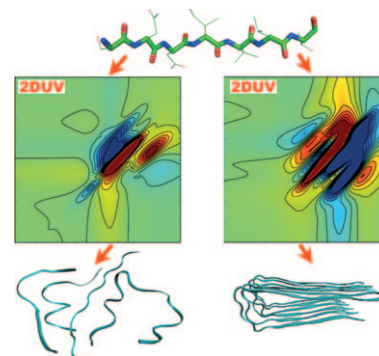
Fibril Formation

J. Jiang, S. Mukamel* — 9666–9669



Two-Dimensional Ultraviolet (2DUV) Spectroscopic Tools for Identifying Fibrillation Propensity of Protein Residue Sequences

Distinctive feature: Two-dimensional ultraviolet (2DUV) spectroscopy can be utilized to identify protein sequences capable of forming fibrils. The Rosetta free energy of proteins, which is a good indicator of fibrillation propensity, has distinct 2DUV signatures. An additional positive diagonal peak at 54 000–58 000 cm^{-1} in chiral $xxxy$ spectra serves as an indicator of the ability of protein sequences to form amyloid-like fibrils.

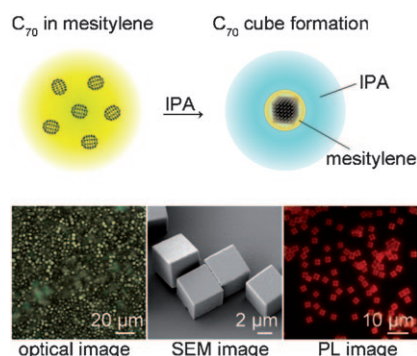


Fullerene Crystals

C. Park, E. Yoon, M. Kawano, T. Joo, H. C. Choi* — 9670–9675



Self-Crystallization of C_{70} Cubes and Remarkable Enhancement of Photoluminescence



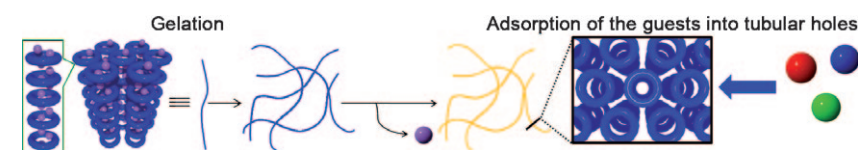
Good solvent, poor solvent: A simple precipitation method enabled the spontaneous formation of homogeneous C_{70} cube crystals by self-crystallization in cavities of a good solvent (mesitylene) surrounded by a poor solvent (isopropyl alcohol, IPA; see picture). The enormously increased photoluminescence (PL) intensity of the C_{70} cube crystals relative to that of C_{70} powder was mainly attributed to the high crystallinity of the cubes.

Supramolecular Gels

T. Nakagaki, A. Harano, Y. Fuchigami, E. Tanaka, S. Kidoaki, T. Okuda, T. Iwanaga, K. Goto, T. Shinmyozu* — 9676–9679



Formation of Nanoporous Fibers by the Self-Assembly of a Pyromellitic Diimide-Based Macrocycle



Spaghetti sensor: A pyromellitic diimide based macrocycle selectively gels N,N -dimethylaniline to form a nanofibril structure. Removal of N,N -dimethylaniline forms channel type empty cavities

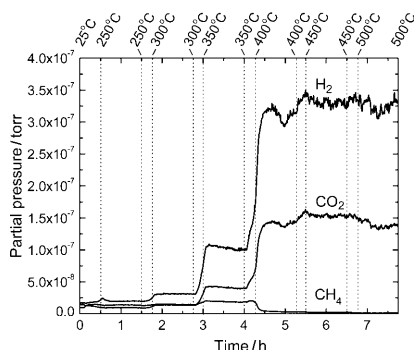
constructed from stacked molecules of the macrocycle in the nanofiber. By taking advantage of host ability of **1**, the nanofibers can serve as a chemosensor for the π -donating compounds.

Heterogeneous Catalysis

G. Zhou, L. Barrio, S. Agnoli, S. D. Senanayake, J. Evans, A. Kubacka, M. Estrella, J. C. Hanson, A. Martínez-Arias, M. Fernández-García, J. A. Rodríguez* — 9680–9684



High Activity of $\text{Ce}_{1-x}\text{Ni}_x\text{O}_{2-y}$ for H_2 Production through Ethanol Steam Reforming: Tuning Catalytic Performance through Metal–Oxide Interactions

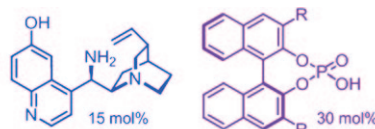


The importance of the oxide: $\text{Ce}_{0.8}\text{Ni}_{0.2}\text{O}_{2-y}$ is an excellent catalyst for ethanol steam reforming (see graph). Metal–oxide interactions perturb the electronic properties of the small particles of metallic nickel present in the catalyst under the reaction conditions and thus suppress any methanation activity. The nickel embedded in ceria induces the formation of O vacancies, which facilitate cleavage of the O–H bonds in ethanol and water.



α Branched leads to γ : The direct and enantioselective γ alkylation of α -substituted α,β -unsaturated aldehydes under dienamine catalysis has been achieved. A cooperative catalysis system that involves dienamine activation of α -branched enals

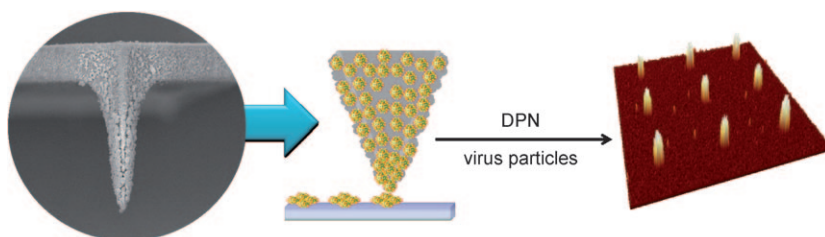
and chiral Brønsted acid catalysis promotes an S_N1 -alkylation pathway while ensuring complete γ -site selectivity and high stereocontrol (see scheme; Bn = benzyl).



Asymmetric Organocatalysis

G. Bergonzini, S. Vera,
P. Melchiorre* _____ **9685–9688**

Cooperative Organocatalysis for the Asymmetric γ Alkylation of α -Branched Enals



Pen-pushing: Direct-write dip-pen nanolithography (DPN) using a tip coated with nanoporous poly(2-methyl-2-oxazoline) allows the creation of precise patterns of large-sized biomaterials such as viruses.

The hydrogel tip absorbs the virus-containing ink solution and atomic force microscopy is used to transport it to a surface (see picture).

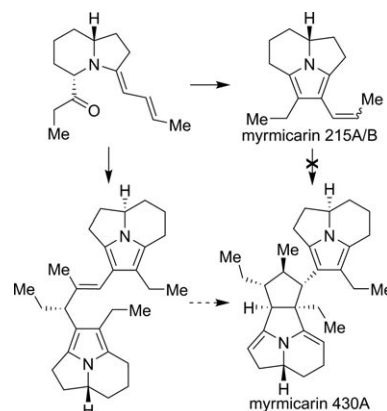
Virus Patterning

Y.-H. Shin, S.-H. Yun, S.-H. Pyo, Y.-S. Lim,
H.-J. Yoon, K.-H. Kim, S.-K. Moon,
S. W. Lee, Y. G. Park, S.-I. Chang,
K.-M. Kim, J.-H. Lim* _____ **9689–9692**

Polymer-Coated Tips for Patterning of Viruses by Dip-Pen Nanolithography



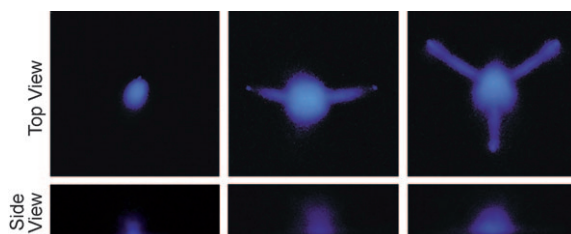
Off to a good start: Use of a carefully designed building block coupled with several highly selective reactions has enabled the syntheses of the monomeric myrmicarins (see scheme) and the investigation of higher-order oligomer synthesis by enabling access to previously unobtainable stereochemical arrangements. These studies, in combination with quantum chemical calculations, question whether the higher-order structures can be obtained through acid-promoted biomimetic synthesis.



Oligomeric Natural Products

S. A. Snyder,* A. M. ElSohly,
F. Kontes _____ **9693–9698**

Synthetic and Theoretical Investigations of Myrmicarins Biosynthesis



It's the pits: Increased efficiency and controllability of sonochemical reactions was achieved with silicon surfaces on which pits were micromachined to entrap gas, which, upon ultrasonic excitation,

emits a stream of microbubbles (see picture). The microbubbles are chemically active at ultrasonic amplitudes well below those necessary for sonochemical activity in conventional reactors.

Sonochemistry

D. Fernandez Rivas,* A. Prosperetti,
A. G. Zijlstra, D. Lohse,
H. J. G. E. Gardeniers _____ **9699–9701**

Efficient Sonochemistry through Microbubbles Generated with Micromachined Surfaces

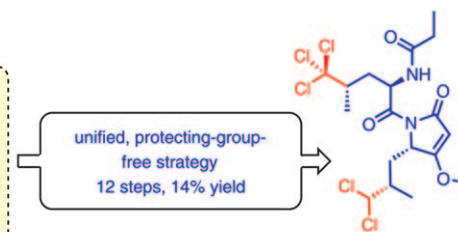
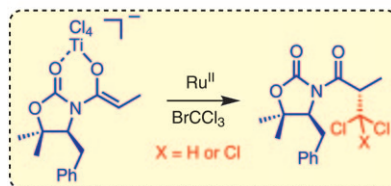


Chlorinated Natural Products

Z. Gu, A. Zakarian* — 9702–9705



Concise Total Synthesis of Sintokamides A, B, and E by a Unified, Protecting-Group-Free Strategy



One for all: A group of polychlorinated marine peptides known as sintokamides show intriguing activity against hormone-refractory prostate cancer cells. Three members of the group have now been

synthesized by a general strategy enabled by a ruthenium-catalyzed radical chloroalkylation of titanium enolates (see scheme).

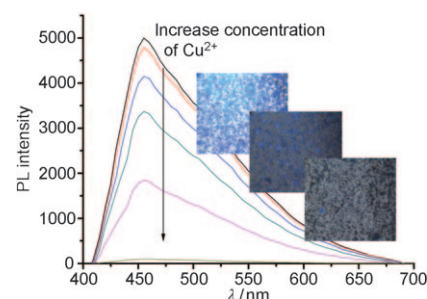
Chemosensors

E. Z. Lee, Y.-S. Jun,* W. H. Hong,*
A. Thomas, M. M. Jin — 9706–9710



Cubic Mesoporous Graphitic Carbon(IV) Nitride: An All-in-One Chemosensor for Selective Optical Sensing of Metal Ions

I can Cu: Cubic mesoporous graphitic carbon nitride (c-mpg-C₃N₄) is an all-in-one chemosensor that plays the roles of ligand, fluorophore, and supporting material, enables the simple detection of metal ions, and is highly selective and sensitive to Cu²⁺.



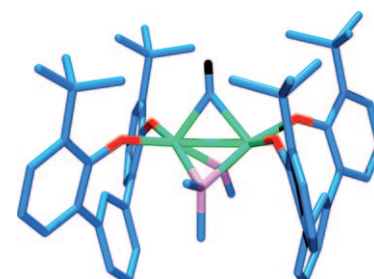
C–N Bond Cleavage

S. Sarkar, J. A. Culver, A. J. Peloquin,
I. Ghiviriga, K. A. Abboud,
A. S. Veige* — 9711–9714



Primary Carbon–Nitrogen Bond Scission and Methyl Dehydrogenation across a W–W Multiple Bond

It takes two: A W–W triple bond provides four electron equivalents to activate an amido C–N bond and dehydrogenate a methyl group to produce a bridging methylidyne W^V–W^V complex (see picture: W green, C blue, N pink, O red, H black). Two detectable intermediates provide insight into this unusual sequence of bond ruptures.

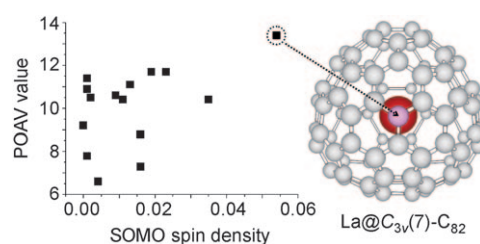


Endohedral Fullerenes

T. Akasaka,* X. Lu, H. Kuga, H. Nikawa,
N. Mizorogi, Z. Slanina, T. Tsuchiya,
K. Yoza, S. Nagase* — 9715–9719

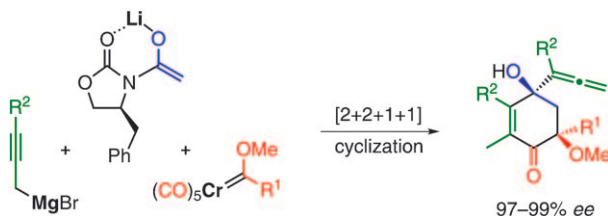


Dichlorophenyl Derivatives of La@C_{3v}(7)-C₈₂: Endohedral Metal Induced Localization of Pyramidalization and Spin on a Triple-Hexagon Junction



Core controls shell: The X-ray structure of a dichlorophenyl adduct of the unprecedented La@C_{3v}(7)-C₈₂ shows that the substituent is singly bonded to a triple-hexagon junction C atom, which has pronounced radical character due to

strong metal–cage interactions, revealed by plotting theoretical p-orbital axis vector (POAV) values against SOMO spin densities for the different types of C atoms (see picture).



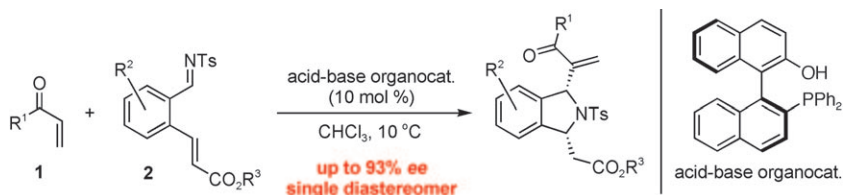
Three metal cooperation promotes the one-pot selective coupling of a chromium carbene complex, an imide lithium enolate, and a propargylic organomagnesium reagent giving access to novel and densely functionalized 4-allenyl-2-cyclohexenones

(see scheme). These useful synthetic intermediates have been prepared through a cyclization process that involves newly reported reaction steps and an unusually high level of asymmetric induction.

Multicomponent Reactions

J. Barluenga,* M. G. Suero,
R. De la Campa, J. Flórez — 9720–9724

Enantioselective Synthesis of 4-Hydroxy-2-cyclohexenones through a Multicomponent Cyclization



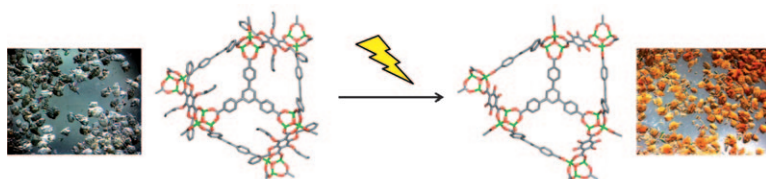
Facile, selective, and organic: Acid–base organocatalyzed aza-Morita–Baylis–Hillman/aza-Michael domino reactions of α,β -unsaturated carbonyl compounds **1** with *N*-tosylimines **2** have been devel-

oped. The enantioselective process produces the highly functionalized isoindoline as a single diastereomer (Ts = 4-toluenesulfonyl).

Organocatalysis

S. Takizawa, N. Inoue, S. Hirata,
H. Sasai* — 9725–9729

Enantioselective Synthesis of Isoindolines: An Organocatalyzed Domino Process Based On the aza-Morita–Baylis–Hillman Reaction



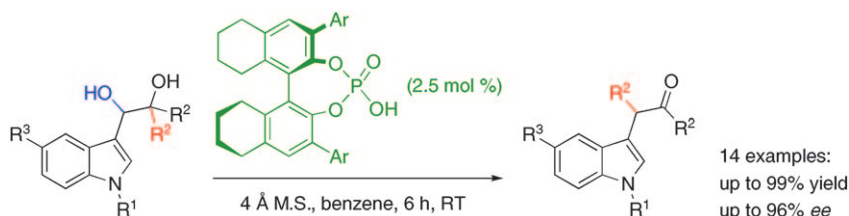
Seeing the light: Two highly porous metal–organic frameworks (MOFs) were transformed using UV light to produce

MOFs with hydroxy and catechol groups through an unusual postsynthetic deprotection reaction (see scheme).

Light-Activated MOFs

K. K. Tanabe, C. A. Allen,
S. M. Cohen* — 9730–9733

Photochemical Activation of a Metal–Organic Framework to Reveal Functionality



Asymmetric pinacol rearrangement: The pinacol rearrangement has long been known to be difficult to control in terms of regioselectivity and stereoselectivity. It has been found that indolyl diols can be

treated with chiral phosphoric acids to effect a regio- and enantioselective pinacol rearrangement with high efficiency (see scheme; Ar = 1-naphthyl).

Organocatalysis

T. Liang, Z. Zhang,
J. C. Antilla* — 9734–9736

Chiral Brønsted Acid Catalyzed Pinacol Rearrangement

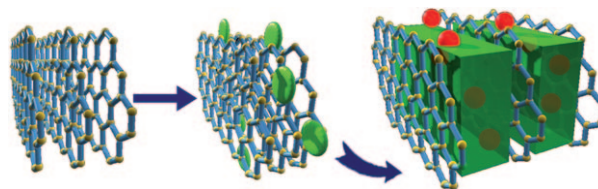


Graphene

Q. Ji,* I. Honma,* S.-M. Paek, M. Akada,
J. P. Hill, A. Vinu, K. Ariga* **9737–9739**



Layer-by-Layer Films of Graphene and Ionic Liquids for Highly Selective Gas Sensing



The best of both worlds: Graphene/ionic liquid (G-IL) layered films were obtained by direct reduction of graphene oxide in the presence of ionic liquids, followed by reassembly through electrostatic layer-by-layer (LbL) adsorption (see picture). The

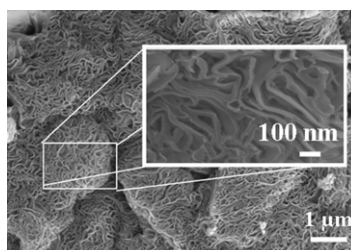
layer spacing of the graphene sheets is regularly expanded upon insertion of ionic liquid molecules (green discs). Selective sensing of aromatic compounds (red spheres) by using the G-IL LbL films was also achieved.

Mesostructures

H. Qi, X. Roy, K. E. Shopsowitz,
J. K.-H. Hui,
M. J. MacLachlan* **9740–9743**



Liquid-Crystal Templating in Ammonia: A Facile Route to Micro- and Mesoporous Metal Nitride/Carbon Composites



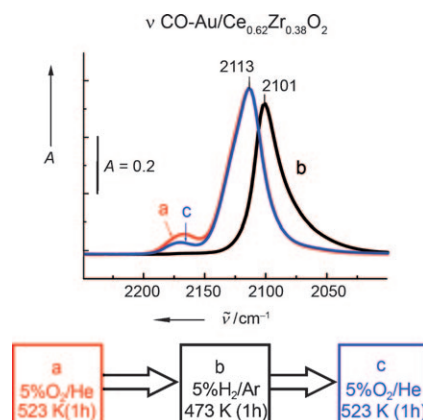
Templating in liquid ammonia: The first porous materials formed by templating in liquid ammonia utilize the liquid-crystalline phase formed by cellulose/NH₄SCN in NH₃. By changing the proportions of reactants, the porosity, surface area, and morphology of the materials can be modified. The as-synthesized mesoporous materials present a lamellar morphology (see picture).

Surface Chemistry

J. M. Cies, E. del Río, M. López-Haro,
J. J. Delgado, G. Blanco, S. Collins,
J. J. Calvino, S. Bernal* **9744–9748**



Fully Reversible Metal Deactivation Effects in Gold/Ceria–Zirconia Catalysts: Role of the Redox State of the Support



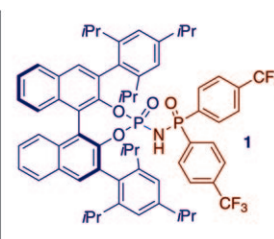
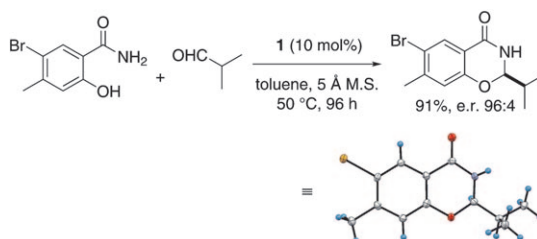
Strong and reversible modification of the chemical properties of supported Au nanoparticles caused by alternating oxidizing (a and c) and reducing (b) pre-treatment of Au/CeO₂–ZrO₂ catalysts were revealed by a methodology that combines FTIR spectroscopy (see picture), studies on the volumetric adsorption of CO and ultimate oxygen storage capacity, determination of metal dispersion by electron microscopy, and X-ray photoelectron spectroscopy.

Organocatalysis

S. Vellalath, I. Čorić, B. List* **9749–9752**

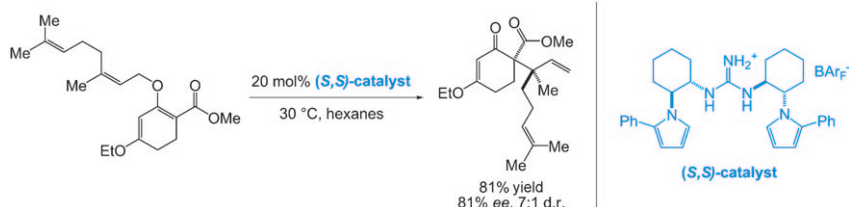


N-Phosphinyl Phosphoramidate—A Chiral Brønsted Acid Motif for the Direct Asymmetric N,O-Acetalization of Aldehydes



Fine-tuning the sites: The readily accessible N-phosphinyl phosphoramidate **1** proved to be highly efficient and enantioselective in catalyzing the title reaction. The synthetic utility of this methodology

was demonstrated with the first catalytic asymmetric synthesis of the analgesic pharmaceutical (R)-chlorothenoxazine (see scheme).



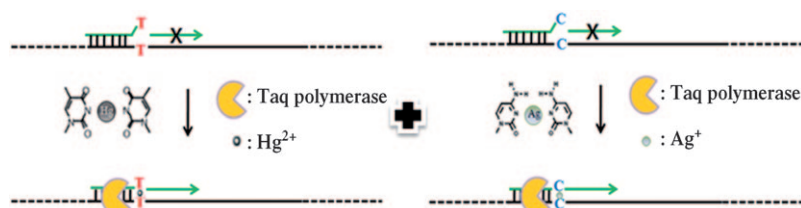
A chiral guanidinium ion is shown to catalyze enantioselective Claisen rearrangements of *O*-allyl β -ketoesters in 78–87% *ee* (see scheme). The pericyclic

nature of the process allows products containing vicinal stereogenic centers to be accessed with both enantio- and diastereocontrol.

Organocatalysis

C. Uyeda, A. R. Rötheli,
E. N. Jacobsen* ————— 9753–9756

Catalytic Enantioselective Claisen
Rearrangements of *O*-Allyl β -Ketoesters



Metal ion, an astonishing illusionist: In a new concept, “illusionary” polymerase activity is intentionally triggered at T–T and C–C mismatched primers by Hg^{2+} and Ag^+ ions, respectively. A novel strategy to construct molecular-scale logic

gates utilizes the nonnatural polymerase activity induced by the metal ions by rational design of the primers and selection of the type of DNA polymerase (see picture).

DNA Technology

K. S. Park, C. Jung,
H. G. Park* ————— 9757–9760

“Illusionary” Polymerase Activity
Triggered by Metal Ions: Use for
Molecular Logic-Gate Operations



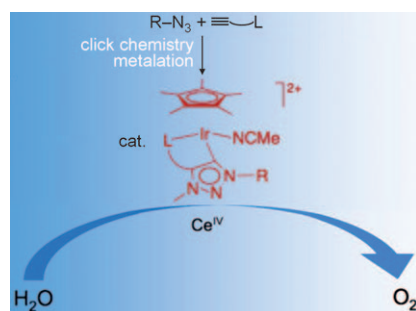
A formal introduction: The conceptually new N-heterocyclic carbene catalyzed formal insertion of arynes into the $\text{C}_{\text{formyl}}\text{--H}$ bond of aldehydes demonstrates the compatibility of nucleophilic NHCs with electrophilic arynes. This NHC-cata-

lyzed hydroacylation of arynes allows the conversion of aliphatic, α,β -unsaturated, and aromatic aldehydes into aryl ketones; 27 examples and a preliminary mechanistic investigation are provided.

Organocatalysis

A. T. Biju, F. Glorius* ————— 9761–9764

Intermolecular N-Heterocyclic Carbene
Catalyzed Hydroacylation of Arynes



Think oxygen: Iridium(III) complexes containing an unusually bound chelating triazolyldiene ligand (see picture) show excellent activity towards water oxidation, producing hundreds of milliliters or O_2 per milligram of iridium. The active catalysts include either an ylide or an unusually bound pyridylidene as the chelating L group and are readily accessible by click chemistry.

O_2 Generation

R. Lalrempuia, N. D. McDaniel,
H. Müller-Bunz, S. Bernhard,*
M. Albrecht* ————— 9765–9768

Water Oxidation Catalyzed by Strong
Carbene-Type Donor-Ligand Complexes of
Iridium

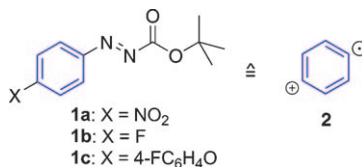


Synthetic Equivalents

S. B. Höfling, A. L. Bartuschat,
M. R. Heinrich* _____ **9769–9772**



4-Substituted *tert*-Butyl
Phenylazocarboxylates—Synthetic
Equivalents for the *para*-Phenyl Radical
Cation



First electrophile, then radical: 4-Substituted *tert*-butyl phenylazocarboxylates **1** are versatile synthetic equivalents of the *para*-phenyl radical cation **2**. The *tert*-butyloxycarbonylazo group enables nucleophilic substitutions to proceed under mild conditions and can later be employed for the generation of aryl radicals.

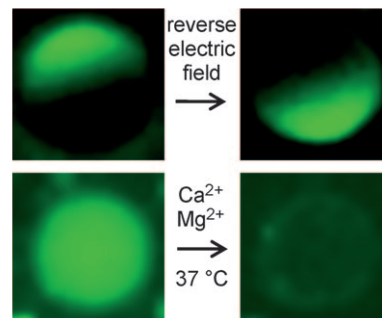
Microcontainers

L. Lin, S. Beyer, T. Wohland, D. Trau,*
D. Lubrich* _____ **9773–9776**



Surface-Bound Microenclosures for
Biomolecules

It's a trap! A simple process based on reverse-phase layer-by-layer encapsulation can be used to produce surface-bound semipermeable microenclosures that can trap biomolecules. Biomolecules such as nucleic acids and proteins can be encapsulated whilst preserving their functionality. Electrophoresis can be used to create sharp concentration gradients, and enzymatic reactions such as DNA digestion can be controlled by diffusion of ions into the microenclosures (see picture).



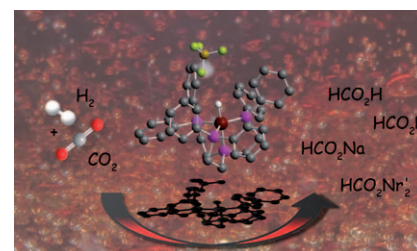
Iron-Catalyzed CO₂ Hydrogenation

C. Federsel, A. Boddien, R. Jackstell,
R. Jennerjahn, P. J. Dyson, R. Scopelliti,
G. Laurenczy,* M. Beller* — **9777–9780**



A Well-Defined Iron Catalyst for the
Reduction of Bicarbonates and Carbon
Dioxide to Formates, Alkyl Formates, and
Formamides

A will of iron: An active well-defined iron complex (see structure; gray C, white H, yellow B, green F, brown Fe, pink P) catalyzes the title reaction (see scheme). The iron-catalyzed reduction of readily available bicarbonates to formates has also been demonstrated for the first time. This reaction could be an important step in the use of CO₂ for hydrogen storage.



Supporting information is available on www.angewandte.org (see article for access details).



A video clip is available as Supporting Information on www.angewandte.org (see article for access details).

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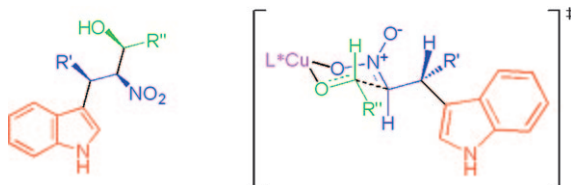
Preview _____ **9785**

The issues for November 2010 appeared online on the following dates

Issue 45: October 26 • Issue 46: November 4 • Issue 47: November 11 • Issue 48: November 17

Corrigendum

In this Communication (DOI: 10.1002/anie.200801373) the Friedel-Crafts/Henry (FCH) adduct with 1*R*,2*S*,3*S* configuration was obtained as the main product in Table 2, entry 6. The relative configuration of this adduct was determined by X-ray crystallographic analysis, but the wrong 1*R**,2*R**,3*R** form was depicted. The correct molecular structure of the main FCH adducts is therefore given below (left). Accordingly, the mechanism of Scheme 2 should be reconstructed. A correct formula for the proposed transition state is shown below (right). The authors apologize for this oversight.



Tandem Catalytic Asymmetric Friedel–Crafts/Henry Reaction: Control of Three Contiguous Acyclic Stereocenters

T. Arai,* N. Yokoyama — 4989–4992

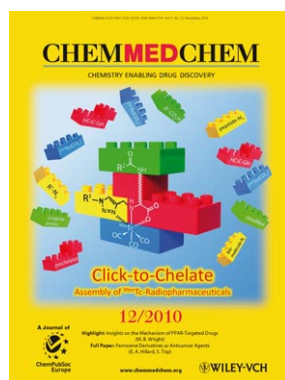
Angew. Chem. Int. Ed. 2008, 47

DOI DOI: 10.1002/anie.200801373

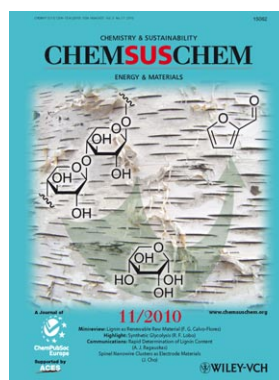
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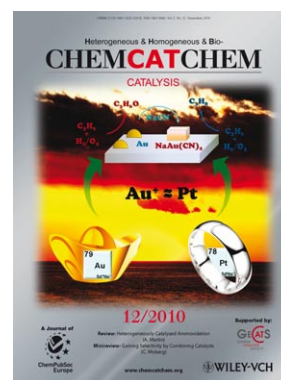
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