



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:

C. Costentin, M. Robert, J. Savéant, C. Tard

Inserting a Hydrogen Bond Relay between Proton Exchanging Sites in Proton-Coupled Electron Transfers

Q. Liu, G. Li, J. He, J. Liu, P. Li, A. Lei*

Palladium-Catalyzed Aerobic Oxidation and Carbonylation of Arylboronate Esters under Mild Conditions

A. C. Filippou,* O. Chernov, K. W. Stumpf, G. Schnakenburg
Metal-Silicon Triple Bonds: The Molybdenum Silylidyne Complex $[\text{Cp}(\text{CO})_2\text{Mo}=\text{SiR}]$

K. Meister, J. Niesel, U. Schatzschneider,* N. Metzler-Nolte,* D. A. Schmidt, M. Havenith*

Metal-Carbonyl Complexes as a Method for Label-Free Live-Cell Imaging by Raman Microspectroscopy

A. Wilbuer, D. H. Vlecken, D. J. Schmitz, K. Kräling, K. Harms, C. P. Bagowski, E. Meggers*

Iridium Complex with Antiangiogenic Properties

A. C. M. Ferreón, C. R. Moran, J. C. Ferreón, A. A. Deniz*

Parkinson's-Related Mutation Alters the α -Synuclein Folding Landscape

J. M. Slattery,* A. Higelin, T. Bayer, I. Krossing*

A Simple Route to Univalent Gallium Salts of Weakly Coordinating Anions

R. Rose, S. Erdmann, S. Bovens, A. Wolf, M. Rose, S. Hennig, H. Waldmann, C. Ottmann*

Identification and Structure of Small-Molecule Stabilizers of 14-3-3 Protein-Protein Interactions

A. Schlossbauer, S. Warncke, P. E. Gramlich, J. Kecht, A. Manetto, T. Carell, T. Bein*

A Programmable DNA-Based Molecular Valve for Colloidal Mesoporous Silica



“If I wasn’t a scientist, I would be an art and antiquities researcher.

The secret of being a successful scientist is to be creative and innovative, to work hard, and most importantly, to be lucky ...”

This and more about Itamar Willner can be found on page 2968.

Author Profile

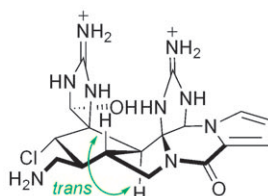
Itamar Willner _____ 2968 – 2970

Molecular Orbitals and Organic Chemical Reactions Ian Fleming

Books

reviewed by A. Kirschning _____ 2971

The marine alkaloid palau’amine has been prepared for the first time in totally synthetic form by a series of cascade and one-pot transformations. These reactions exploit the inherent reactivity of functional groups and deliver interesting solutions to daunting synthetic challenges.



Highlights

Natural Product Synthesis

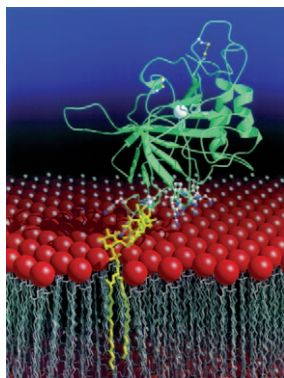
H. J. Jessen, K. Gademann* 2972 – 2974

Total Synthesis of the Marine Alkaloid Palau’amine

Carbonic Anhydrase

A. Dunkel, T. Hofmann* — 2975–2977

Carbonic Anhydrase IV Mediates the Fizz of Carbonated Beverages



Sensational findings: The fizzy taste sensation we perceive when we consume carbonated beverages is mediated by carbonic anhydrase IV, an enzyme catalyzing the conversion of carbon dioxide to hydrogen carbonate and free protons. The picture shows the enzyme (green) linked to a membrane through a glycosylphosphatidylinositol tail (yellow). The zinc ion of the active site is represented by a white sphere.

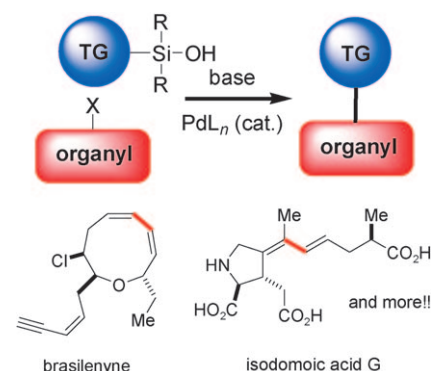
Minireviews

Organosilicon Chemistry

S. E. Denmark,* J. H.-C. Liu — 2978–2986

Silicon-Based Cross-Coupling Reactions in the Total Synthesis of Natural Products

Versatile but underutilized: Transition-metal-catalyzed cross-coupling reactions have revolutionized the synthesis of complex organic molecules. Recent advances in organosilicon chemistry have significantly expanded the utility of these coupling partners. This Minireview presents case studies wherein silicon-based cross-coupling reactions play a strategic role in the total synthesis of selected natural products (see scheme; TG = transferable group).

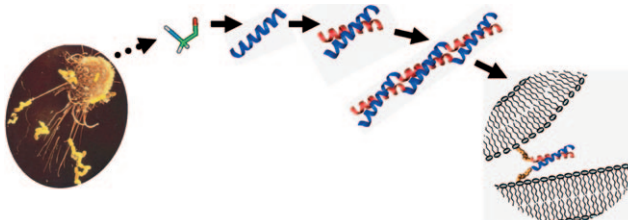


Reviews

Coiled Coils

H. Robson Marsden,
A. Kros* — 2988–3005

Self-Assembly of Coiled Coils in Synthetic Biology: Inspiration and Progress



Cues from nature: The coiled-coil motif is a simple molecular building block ubiquitous in nature that has an impressive range of functions, with roles in protein binding, the formation of structural edifi-

ces, and dynamic processes. These functions inspire research in synthetic biology, in which the coiled-coil motif is utilized in functional units, assemblies, and systems.

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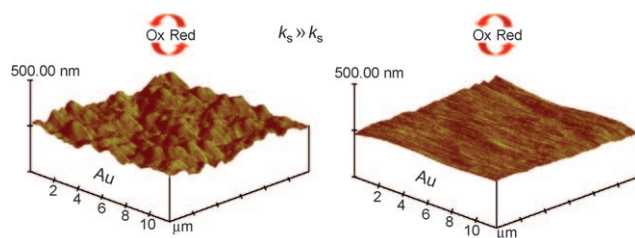
Communications

Electrochemistry



A. M. Nowicka,* U. Hasse, G. Sievers,
M. Donten, Z. Stojek, S. Fletcher,
F. Scholz* _____ 3006–3009

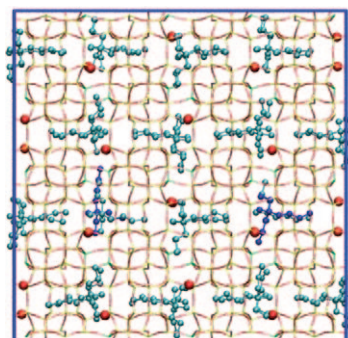
Selective Knockout of Gold Active Sites



It's a knockout! When the surface of gold electrodes is treated with hydroxyl radicals, the electron-transfer rates k_s of electrochemical reactions involving radical intermediates are drastically reduced

(see picture). This observation can be explained by the selective knockout of gold active sites associated with partially filled d orbitals.

Minority excluded: If mixtures of chiral compounds are adsorbed in Al-substituted MFI zeolite containing nonframework cations, a process occurs that ultimately might be used for enantioseparation. If one enantiomer is present in excess it will organize the nearby cations, which in turn influence all other cations through long-range interaction, such that all adsorption sites become effectively chiral. This suppresses the minority enantiomer.



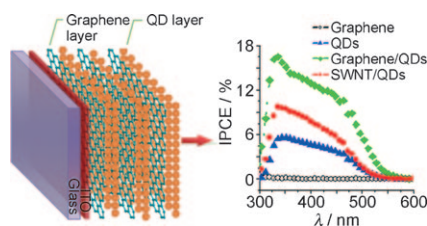
Enantioseparation

T. S. van Erp,* T. P. Caremans,
D. Dubbeldam, A. Martin-Calvo, S. Calero,
J. A. Martens _____ 3010–3013

Enantioselective Adsorption in Achiral Zeolites



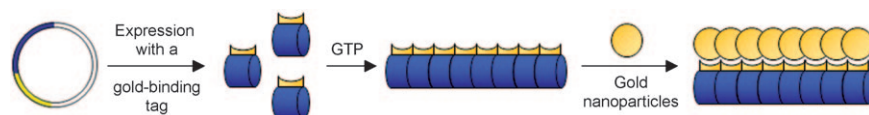
Layering in the sun: Layered graphene and quantum dot (QD) films can be fabricated simply on transparent conducting indium tin oxide (ITO) substrates from aqueous solutions. The structure and favorable work function of graphene make it effective for the collection and transfer of photogenerated charges to the electrode, resulting in a high-performance photovoltaic device (see picture; IPCE = incident photon-to-charge-carrier conversion efficiency, SWNT = single-walled carbon nanotube).



Photovoltaics

C. X. Guo, H. B. Yang, Z. M. Sheng,
Z. S. Lu, Q. L. Song,
C. M. Li* _____ 3014–3017

Layered Graphene/Quantum Dots for Photovoltaic Devices



Naturally born nanowires: The spontaneous self-assembly of the bacterial cytoskeletal protein FtsZ was used to position metal nanoparticles in an orderly fashion to fabricate a metal nanowire (see scheme). The genetic fusion of each

monomer with an appropriate short protein motif enables the specific binding not only of nanoparticles, but also of other inorganic particles and biological molecules.

Nanotechnology

N. Ostrov, E. Gazit* _____ 3018–3021

Genetic Engineering of Biomolecular Scaffolds for the Fabrication of Organic and Metallic Nanowires



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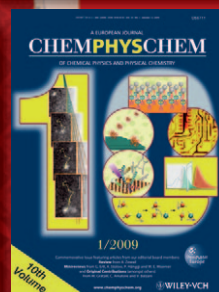


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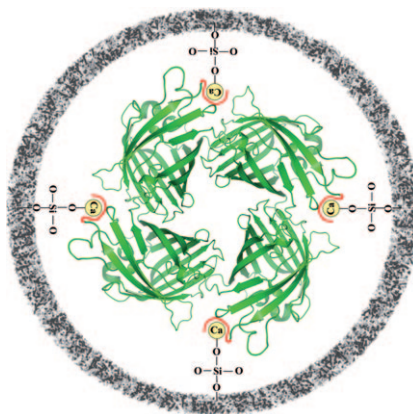
Topics

catalysis, biochemical imaging,
chemical biology, bionanotechnology,
proteomics, spectroscopy, solar cells



WILEY-VCH

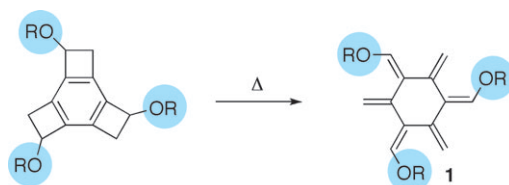
Si'l vous plait? A facile and general method has been developed to encapsulate polyhistidine-tagged proteins in silica nanoparticles (NPs; gray, see picture) using calcium ions (yellow). The enhanced green fluorescent protein (EGFP) encapsulated in the silica NPs shows a substantial increase in fluorescence intensity and stability against denaturants, protease, and heat.



Protein Encapsulation

A. Cao,* Z. Ye, Z. Cai, E. Dong, X. Yang, G. Liu, X. Deng, Y. Wang, S.-T. Yang, H. Wang,* M. Wu, Y. Liu — **3022–3025**

A Facile Method To Encapsulate Proteins in Silica Nanoparticles: Encapsulated Green Fluorescent Protein as a Robust Fluorescence Probe



You rang m'lord? Thermally induced successive ring openings of tris(alkoxytricyclobutabenzene) to hexaradialene are described. The isomerization reaction was highly stereoselective to afford a stereo-

defined hexaradialene **1**, given that the hydroxy groups on the four-membered rings were protected by bulky substituents.

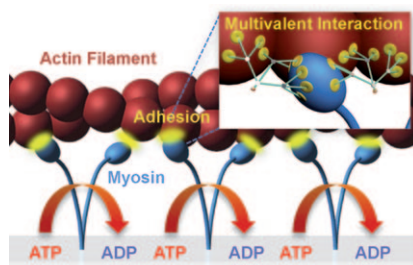
Ring Opening Reactions

S. Shinozaki, T. Hamura, Y. Ibusuki, K. Fujii, H. Uekusa, K. Suzuki* — **3026–3029**

Hexaradialenes by Successive Ring Openings of Tris(alkoxy-tricyclobutabenzene)s: Synthesis and Characterization



A sticky end: A heterotropic conjugate of actin and myosin–actomyosin—is stabilized by a water-soluble dendritic “molecular glue” bearing nine pendant guanidinium ions (see picture; ATP = adenosine triphosphate, ADP = adenosine diphosphate). The ATP-driven sliding motion of actin filaments on a myosin-functionalized coverslip is decelerated or totally arrested with a higher-generation dendrimer, but not with a lower-generation one.



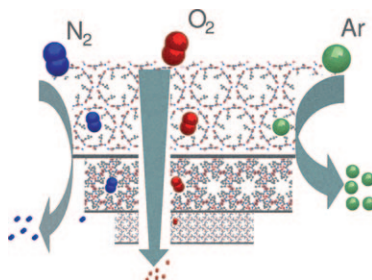
Molecular Motion

K. Okuro, K. Kinbara,* K. Takeda, Y. Inoue, A. Ishijima, T. Aida* — **3030–3033**

Adhesion Effects of a Guanidinium Ion Appended Dendritic “Molecular Glue” on the ATP-Driven Sliding Motion of Actomyosin



Selective pores: Microporous dipeptide crystals can be used as single-crystal membranes. In permeation experiments with N₂, O₂, and Ar, the main components of air, the crystals show high permeabilities and excellent selectivities (see picture). L-Alanyl-L-alanine crystals exhibit extremely high O₂/N₂ selectivities.



Permselective Crystals

R. V. Afonso, J. Durão, A. Mendes, A. M. Damas, L. Gales* — **3034–3036**

Dipeptide Crystals as Excellent Permselective Materials: Sequential Exclusion of Argon, Nitrogen, and Oxygen

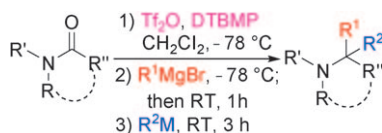


Synthetic Methods

K.-J. Xiao, J.-M. Luo, K.-Y. Ye, Y. Wang,
P.-Q. Huang* 3037–3040



Direct, One-pot Sequential Reductive Alkylation of Lactams/Amides with Grignard and Organolithium Reagents through Lactam/Amide Activation



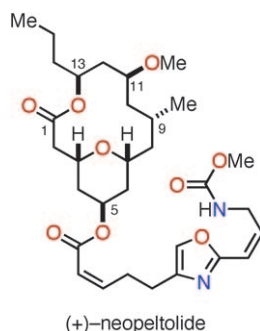
Be dazzled by the sequence: The first efficient and general one-pot method for the reductive bisalkylation of lactams/amides with Grignard and organolithium reagents has been developed (see scheme; DTBMP = 2,6-di-*tert*-butyl-4-methylpyridine, Tf = trifluoromethanesulfonyl).

Natural Product Synthesis

H. Fuwa,* A. Saito,
M. Sasaki 3041–3044



A Concise Total Synthesis of (+)-Neopeltolide



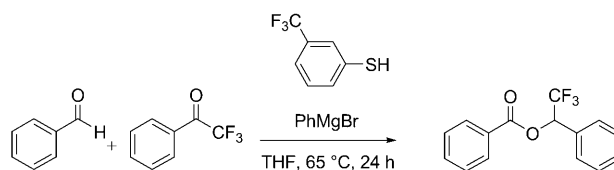
Short and sweet: The highly potent antiproliferative marine macrolide (+)-neopeltolide has been synthesized based on the strategic application of olefin metathesis reactions. The total synthesis proceeded in only 13 steps from commercially available starting materials, and represents the shortest synthesis of (+)-neopeltolide reported to date.

Synthetic Methods

L. Cronin, F. Manoni, C. J. O' Connor,
S. J. Connon* 3045–3048



Tunable Bromomagnesium Thiolate Tishchenko Reaction Catalysts: Intermolecular Aldehyde–Trifluoromethylketone Coupling



Applied knowledge: A new thiolate-catalyzed Tishchenko reaction inspired by biochemical processes features exceptionally good control over the catalyst's steric/electronic properties. The isolation of reaction intermediates provided valua-

ble insights into the reaction mechanism, which in turn has allowed the development of the first cross-Tishchenko coupling reactions between aldehydes and activated ketones (see example).

Stereocontrolled Glycosylation

D. Crich,* C. Navuluri 3049–3052

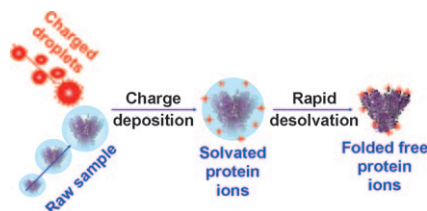


Stereoselective Synthesis of α -Keto-deoxy-D-glycero-D-galacto-nonulosonic Acid Glycosides by Means of the 4,5-O-Carbonate Protecting Group



Unrivaled: A 1-adamantyl thioglycoside derivative of the nonulosonic acid KDN, carrying a 4,5-O-carbonate protecting group, is a highly efficient and α -selective KDN donor when activated using N-

iodosuccinimide (NIS) and trifluoromethanesulfonic acid (TfOH). Glycosylations conducted with this protecting group do not suffer from competing glycal formation.

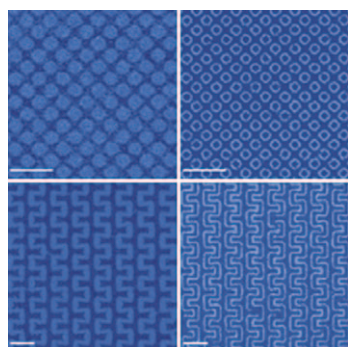


“Soft” ionization: EESI deposits charges on native proteins, which are separated from any high electric field. In this way native proteins can be characterized by mass spectrometry even from raw biological samples without significant conformational changes or reduction in activity. EESI-MS is a sensitive tool for the high-throughput analysis of trace levels of proteins under the native conditions.

Analytical Methods

H. Chen,* S. Yang, M. Li, B. Hu, J. Li, J. Wang — 3053–3056

Sensitive Detection of Native Proteins Using Extractive Electrospray Ionization Mass Spectrometry

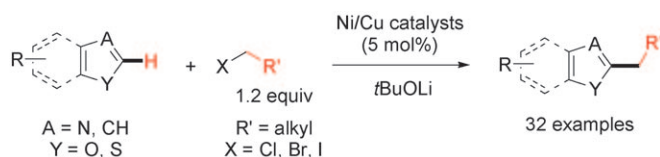


There's more behind these masks: Masks composed of Au nanohole-array films on a poly(dimethylsiloxane) (PDMS) slab were used to create high-density molecular patterns. Nanohole films of different thicknesses produced either edge patterns or 1:1 patterns (see examples; scale bars: 2 μm). Hierarchical patterns were created with a single nanostencil mask by using a patterned PDMS slab or by combining nanostencil printing with microcontact printing.

Nanostencil Printing

M. H. Lee, J. Y. Lin, T. W. Odom* — 3057–3060

Large-Area Nanocontact Printing with Metallic Nanostencil Masks



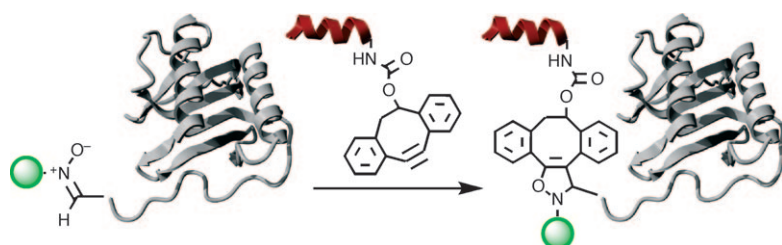
Too selective: A general and straightforward protocol for the cross-coupling of non-activated alkyl halides with heterocyclic C–H bonds has been developed. The transformation is chemo- and regioselective and many functional groups on both

coupling partners are tolerated. The method employs cheap nickel/copper catalysts, and expands significantly the scope of C–H functionalization (see scheme).

C–H Activation

O. Vechorkin, V. Proust, X. L. Hu* — 3061–3064

The Nickel/Copper-Catalyzed Direct Alkylation of Heterocyclic C–H Bonds



Quicker and slicker: An efficient metal-free 1,3-dipolar cycloaddition of dibenzocyclooctynes with nitrones proceeded with rate constants of up to 39 $\text{M}^{-1}\text{s}^{-1}$, or up to

300 times faster than similar reactions with azides. This strategy is useful for the site-specific N-terminal modification of peptides and proteins (see scheme).

Bioorthogonal Reactions

X. Ning, R. P. Temming, J. Dommerholt, J. Guo, D. B. Ania, M. F. Debets, M. A. Wolfert, G.-J. Boons,* F. L. van Delft* — 3065–3068

Protein Modification by Strain-Promoted Alkyne–Nitron Cycloaddition



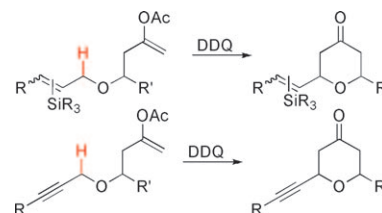
C–H Activation

L. Liu, P. E. Floreancig* — 3069–3072



Structurally and Stereochemically Diverse Tetrahydropyran Synthesis through Oxidative C–H Bond Activation

Simple as pyran: Vinylsilane-substituted tetrahydropyrans, readily accessed through the oxidative C–H bond functionalization of silylallylic or propargylic ethers (see scheme; DDQ = 2,3-dichloro-5,6-dicyano-1,4-benzoquinone), are versatile substrates for a wide range of functional group interconversions and stereocontrolled additions. These processes have applications in convergent target- or diversity-oriented synthesis strategies.

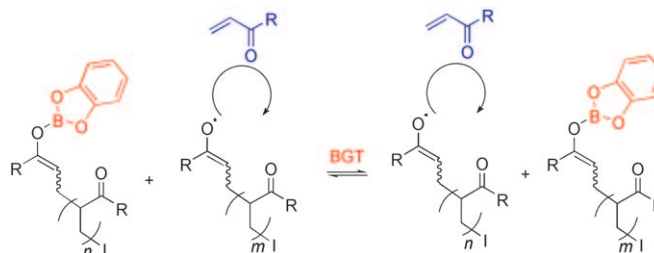


Controlled Radical Polymerization

K. Uehara, C. B. Wagner, T. Vogler, H. Luftmann, A. Studer* — 3073–3076



Poly(vinyl ketone)s by Controlled Boron Group Transfer Polymerization (BGTP)



Boron can do it! Readily prepared catecholboron enolates act as regulators in the controlled radical polymerization of alkyl and aryl vinyl ketones. Control is achieved by an unprecedented reversible

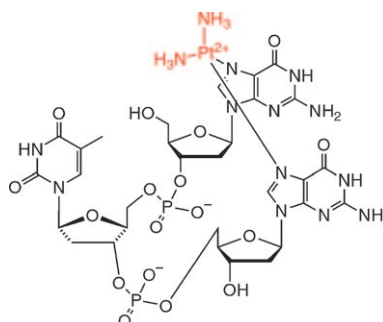
radical boron group transfer (BGT) between enoyl radicals (see scheme). Polymers with $M_n \leq 14\,000 \text{ g mol}^{-1}$ and a polydispersity index of 1.3 can be obtained by this process.

Cisplatin

T. Reißner, S. Schneider, S. Schorr, T. Carell* — 3077–3080



Crystal Structure of a Cisplatin–(1,3-GTG) Cross-Link within DNA Polymerase η



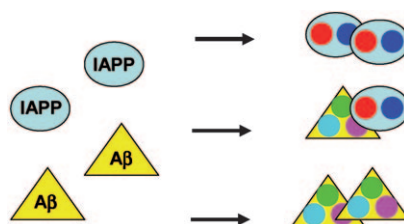
Flipped out: The cisplatin–(1,3-GTG) adduct, a strongly cytotoxic DNA lesion generated by the action of cisplatin therapeutics, is partially bypassed by Y-family DNA polymerase η . Crystal structure data on this DNA–protein complex explain the partial-bypass process. The central dT unit of the lesion is turned out and blocks the movement of the enzyme along the DNA duplex.

Protein Interactions

E. Andreetto, L.-M. Yan, M. Tatarek-Nossol, A. Velkova, R. Frank, A. Kapurniotu* — 3081–3085



Identification of Hot Regions of the A β –IAPP Interaction Interface as High-Affinity Binding Sites in both Cross- and Self-Association



Short peptide sequences are identified as hot regions of the cross-interaction interface of the Alzheimer's disease β -amyloid peptide (A β) with the type 2 diabetes islet amyloid polypeptide (IAPP). They are shown to be high-affinity ligands of both A β and IAPP, thus suggesting common molecular recognition features in amyloid self- and cross-amyloid hetero-assembly.



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



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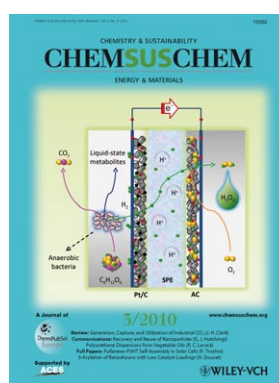
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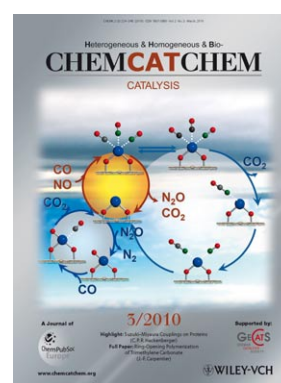
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