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Unifying Concepts in Catalyis

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unicat
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I. Research at a glance

Current challenges in catalysis range from the efficient exploitation of energy resources to the creative use of natural and artificial enzymes.

Our long-term goal is to build a reliable knowledge base across the scientific disciplines for the design and synthesis of new active materials. We want to interconnect the different perceptions and strengths of heterogeneous, homogeneous and biological catalysis.

The key objective is to identify catalytic concepts based on a molecular understanding of the systems and utilise this for the development of novel and more efficient catalysts to be transferred to technological applications.

Significant research targets are:

- **Oxidative coupling of methane into ethene and related compounds.**
  Natural gas consists of up to 98% methane. The development and improvement of processes for the selective activation and conversion of methane is one of the enormous challenges in fundamental and applied chemistry. If methane could be chemically converted into value-added substances, this would provide a significant time buffer before the inevitable departure from the use of oil. This would make the transition to renewable energy sources significantly easier.

- **Biological hydrogen production and hydrogen-driven fuel cells.**
  Molecular hydrogen is one of the most environmentally friendly sources of energy. Based on fundamental findings from basic research, UniCat is developing new strategies for producing hydrogen using light, water and enzymes. The reverse reaction, the generation of electricity from hydrogen and oxygen in a biological fuel cell, is also studied.

- **Development of novel antibiotics.**
  There is an urgent need for new bioactive compounds in the drug development processes of the chemical industry, due for instance to growing resistances to antibiotics. UniCat is searching for alternatives, exploiting the biosynthetic potential of bacteria and fungi.

To study these three main research targets (see above), our highly interdisciplinary research programme is organised into three cross-linked Research Areas. Each Area is divided into several Research Fields.

A) Bridging the materials gap in complex catalysis.

B) “Intelligent” natural and artificial enzymes.

C) Complex reaction engineering.

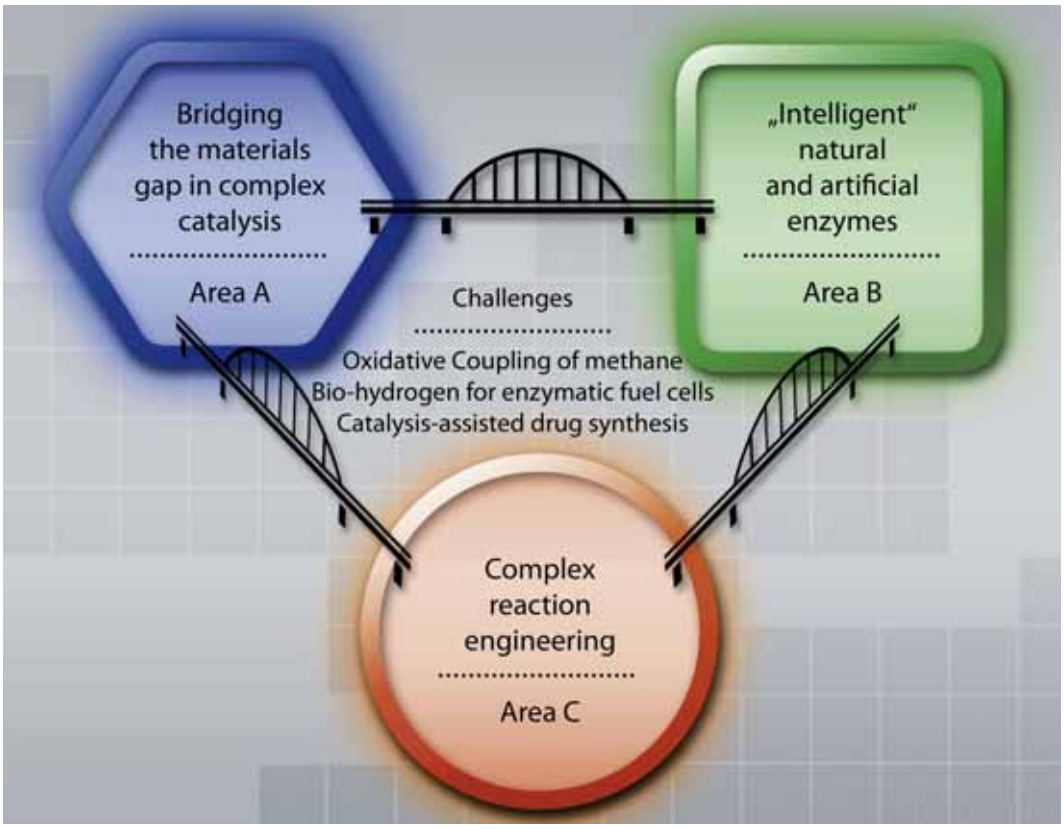


Fig. 1: Research Areas of UniCat.

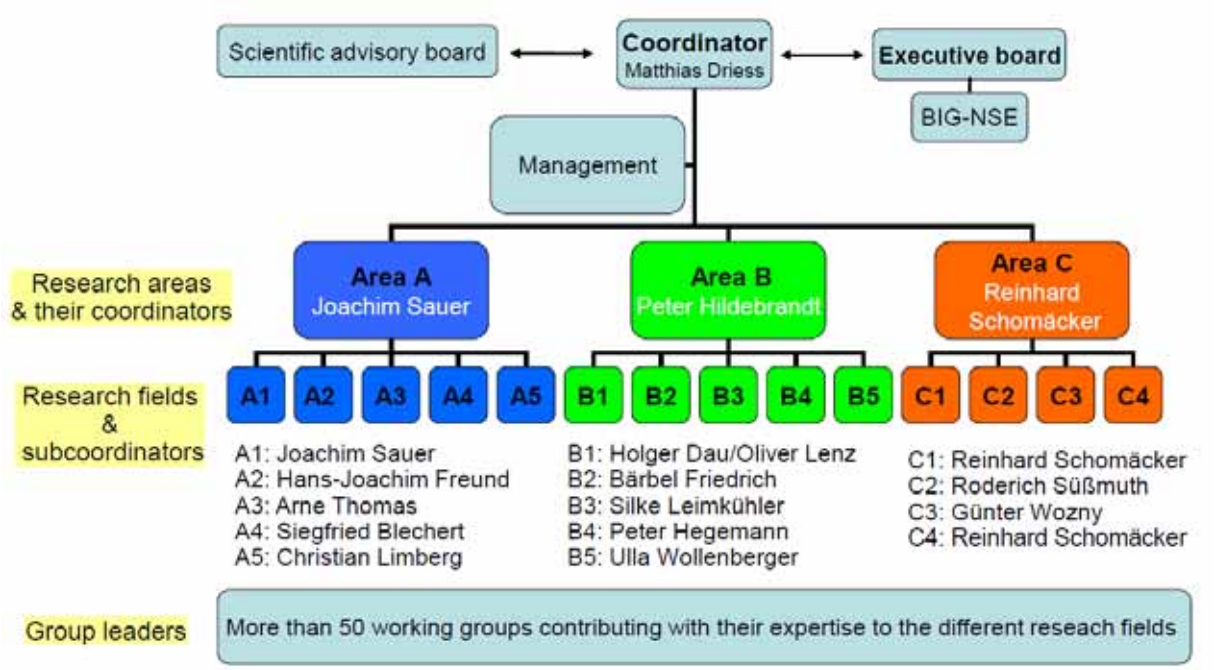
In **Area A**, the central challenge is to explore how the performance of catalytic materials varies depending on different length scales and in different molecular environments.

In **Area B**, the mechanisms of biocatalytic processes of intriguing redox-, light- and voltage-"powered" enzymes are investigated at molecular and cellular levels. The knowledge accumulated in A and B constitutes the basis for improved and novel tailor-made catalysts for technological applications such as energy-saving chemical processes, bio-renewable energy resources and the efficient synthesis of novel drugs.

In **Area C**, the novel catalytic systems developed in Areas A and B will be appropriately scaled up and tested on the mini-plant level, thereby developing novel reactor types and methodologies for process design.

The UniCat consortium provides a wide variety of state-of-the-art experimental and theoretical methods, which are crucial for its success.

II. Organisation



Area A: Bridging the materials gap in complex catalysis

A1: Bridging model systems to real catalysis: oxidative methane coupling
A2: Tailoring metal-support interactions
A3: Hierarchically organised solid catalysts
A4: Consecutive catalysis for fine chemical synthesis
A5: Linking homogeneous and biocatalysis

Area B: "Intelligent" natural and artificial enzymes

B1: Photosynthetic water oxidation, light driven H_2 production, hydrogenase-based biofuel cells
B2: Structure-function analysis of oxygen tolerant hydrogenases
B3: Cofactor insertion and functional investigations on complex molybdoenzymes
B4: Light- and voltage-gated enzymes
B5: Bioelectronic building blocks

Area C: Complex reaction engineering

C1: Processing of solid catalysts
C2: Biocatalysis and process techniques
C3: Process simulation with hierarchic models
C4: Design of integrated catalytic processes

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III. Graduate School BIG-NSE

The **B**erlin **I**nternational **G**raduate School of **N**atural **S**ciences and **E**ngineering (BIG-NSE) is part of UniCat and was founded in 2007 at the Technische Universität Berlin.

The high-level research in catalysis undertaken within the Graduate School covers a broad range of topics, from natural sciences to engineering. The success of this research programme, which is unique in Germany, requires highly-qualified, motivated young researchers for different projects. To this end, the BIG-NSE offers a structured curriculum for obtaining a doctorate within three years.

Curriculum

Students admitted to the School first pass through an “Initial Phase”, usually of three months, to integrate the different branches of science and engineering and establish a common scientific language.

The students not only attend the same lectures and courses, but also work together in the same offices during this Initial Phase. For integration of the students into the School, each new student will have a mentor from the group of previous students of the School. Each student will prepare a research proposal as a guideline for his/her future mentoring. In a final workshop, each student will present his/her project in front of the UniCat Cluster faculty.

Subsequently, the students join their research groups for their individual research project. Annual reports and frequent meetings with their Advisory Committee provide guidelines for the successful continuation of their work. Parallel to their research work, the PhD students attend “Lecture Courses” by members of the UniCat Cluster. The programme of the School includes Summer and Winter Schools that are open to external researchers and students to get develop new ideas, and transfer knowledge to research partners and industry. Training of “soft” skills (e.g. presentations, project management) and excursions to industrial enterprises are offered regularly.

Requirements

The entry requirements for the BIG-NSE are:

- A Master‘s degree or German Diploma in chemistry, biology, physics or engineering.
- A Certificate of English Proficiency (TOEFL with a minimum of 550, or equivalent) for applicants whose native language is neither English nor German.
- Two letters of recommendation.

Recruitment

A central part of the BIG-NSE activities is the recruitment of excellent national and international students that meet the highest international standards of qualification. About 20 highly-qualified students will be recruited every year.

The courses of BIG-NSE start on October 1 each year having following deadlines:

- **MARCH 15** (*Application for a scholarship funded by BIG-NSE*)
- **AUGUST 31** (*Application for attending the School*)

In 2011 a new Master‘s Programme starts. The Master of Science in Catalysis will be offered by all four Universities participating in UniCat. It will be open for students having a BSc in chemistry, physics, biology or engineering. Degree holders in catalysis will attend the BIG-NSE PhD programme, too.

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IV. Research Area A: Bridging the materials gap in complex catalysis

Coordinator

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The activation of small molecules is a key problem in the utilisation of resources for matter and energy conversion in the future. In order to achieve sustainable development, chemical technologies have to be developed in which the activation process is done by selective and (energy-) efficient transformations into a broad range of value-added products with desirable functionalities.

Structure of Research Area A

In Research Area A, structure-property-function relationships of catalytic systems are investigated to elucidate the interplay between the reactivity of catalytic sites and the molecular environment.

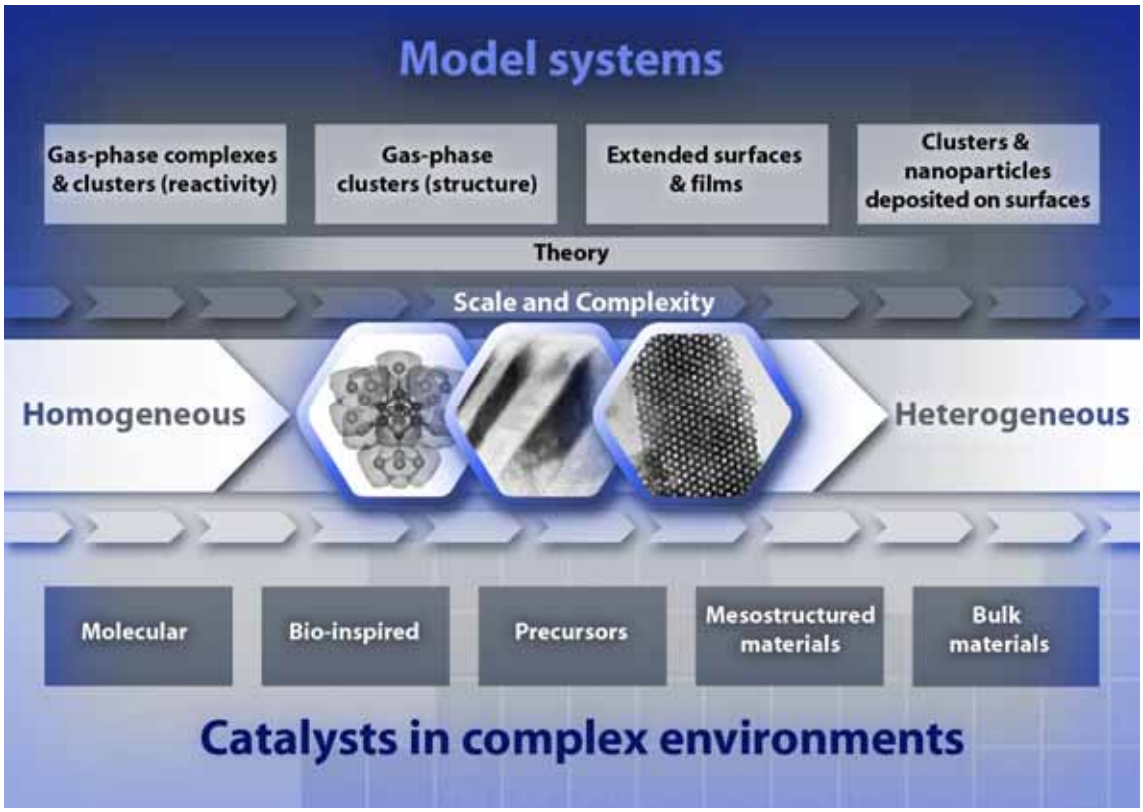

Fig. 2: Research structure in Area A indicating the hierarchical approach to complex catalytic systems.

This Area is subdivided into two parts.

- Research Fields A1 and A2 work on model systems, including “isolated” active centres in the gas phase and on surfaces, as well as metal clusters of different sizes. These systems are analysed with respect to the evolution of properties for metals and metal oxides at different levels of aggregation, ranging from single atoms and molecular assemblies to the solid material.

- Research Fields A3, A4 and A5 deal with catalysts in complex environments and aim to develop new catalysts and novel catalytic routes in synthetic chemistry. Hierarchically organised solid-state catalysts are designed for bridging the materials gap, focussing in particular on metastable MoO_3 and Li-MgO phases for methane conversion. This research is closely linked to a scaling-up and optimisation of catalytic processes for the oxidative coupling of methane in Area C.

Research goals

A comprehensive understanding of catalytic mechanisms in hierarchically organised environments for:

- the design and synthesis of new active materials for heterogeneous and homogeneous catalysis
- the activation of small molecules, e.g. methane, hydrogen
- the transformation of small molecules into value-added products, e.g. methanol, ethylene

Research Fields of Area A

Research Area A is organised into five Research Fields.

A1: Bridging model systems to real catalysis: oxidative methane coupling

A2: Tailoring metal-support interactions

A3: Hierarchically organised solid catalysts

A4: Consecutive catalysis for fine chemical synthesis

A5: Linking homogeneous and biocatalysis

A1: Bridging model systems to real catalysis: oxidative methane coupling

Sub-Coordinator

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State of the art

The direct oxidative coupling of methane (OCM) to C2-hydrocarbons (ethane and ethene) is a reaction of high interest, as it could greatly enhance the sustainability of the petrochemical feedstock for energy and matter conversion. However, this high-temperature process (T = 700-900°C) is characterised by low yield and poor selectivity towards ethene, thereby preventing its economical use at present. However, profitability will change drastically concomitant with the inevitable shortage of non-renewable resources. In addition, the current wasteful practice of flaring methane accounts for some 20% of the total greenhouse gas emissions. Rational improvements to the OCM process are currently limited due to the fact that key steps of the overall mechanism involving heterogeneous and homogeneous (gas-phase) reactions are not understood.

Research goals

A joint effort to understand the mechanistic details of the process and to design and synthese tailored catalysts is essential for a breakthrough in developing an industrial catalytic process (long-term objective) which is as yet unknown. In collaboration with Area C (C3), a complete kinetic model for the full range of process conditions and relevant reactor types will be established that is based on reliable information concerning elementary processes on the molecular scale. Our short-term objective is therefore a detailed understanding of individual steps of this complex reaction with the most important catalyst (Li-doped MgO). This system will be used as a reference that allows a comprehensive methodological approach to be established, something which has so far not been achieved. The idea is to combine experimental and theoretical model studies performed on well-defined surfaces and on gas-phase systems with in-situ studies at ambient pressure. In addition, research into catalysts in various reactor types is necessary (collaboration with C4).

Project team of A1

Prof. Dr. Matthias Driess (TU Berlin)	Metalorganic chemistry, Precursor synthesis, Molecular models for heterogeneous catalysts
Prof. Dr. Hans-Joachim Freund (FHI)	Surface science, thin films on supports
Prof. Dr. Gerard Meijer (FHI)	Infrared (IR) photodissociation spectroscopy
Dr. Karsten Reuter (FHI)	DFT, first principles statistical mechanics and kinetic Monte Carlo
Prof. Dr. Joachim Sauer (HU Berlin)	DFT and ab initio methods, Hybrid QM/QM methods, chemical reactivity
PD Dr. Marek Sierka (HU Berlin)	Genetic algorithms for gas phase clusters and surfaces, DFT method development
Prof. Dr. Matthias Scheffler (FHI)	Electronic structure theory, density-functional theory, first-principles statistical mechanics
Prof. Dr. Robert Schlögl (FHI)	Molecular beam mass spectrometry
Prof. Dr. Martin Wolf (FU Berlin)	Ultrafast laser spectroscopy of surface reaction dynamics
Prof. Dr. Klaus-Peter Dinse (FU Berlin)	High-Frequency EPR spectroscopy

A2: Tailoring metal support interactions

Sub-Coordinator

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State of the art

Surface-rich transition metal particles (≈ 3-8 nm diameter) supported on metal oxides represent the largest class of solid catalysts used in industrial processes. Particle growth of active particles into larger, less active aggregates is regarded to be a major, inevitable problem. Aggregation driven by Ostwald ripening can be influenced by the strength of the metal-support interaction. However, it is known that interactions with small inorganic ligands, e.g. Cl^-, OH^-, can facilitate high degrees of dispersion. On an industrial scale, acetate and citrate ligands have been used in this fashion. Yet the concept of using molecular auxiliaries to redisperse metallic catalysts, despite its attractiveness, is far from being understood or established. Furthermore, the competition between intentionally added ligands and the unintentionally present terminating groups from the support opens up the possibility of the rational design of metal-support phenomena, if the strength and specificity of the interaction between metal nanoparticles and ligand can be tailored.

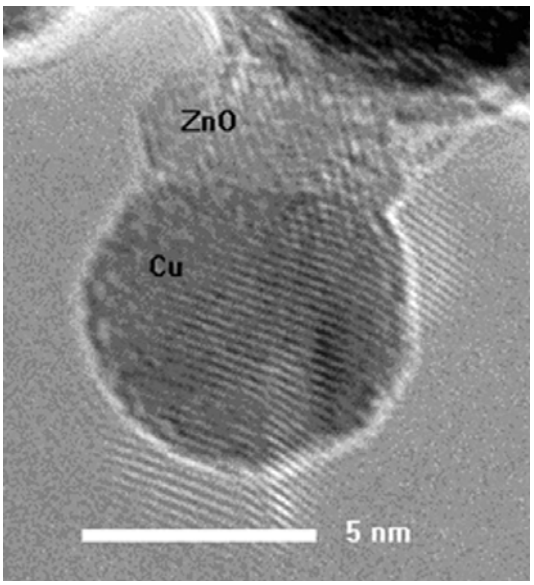

Fig. 3: Transmission electron microscopy (TEM) of an active Cu nanoparticle prepared on ZnO/Alumina support.

Research goals

Information on strength, type and specificity of the interactions with appropriate ligands is being obtained by combining differently scaled model systems similar to the approach introduced in Research Field A1. We are investigating how the presence of such ligands affects the morphology, stability, mobility and reactivity of metal clusters on oxide surfaces and in the gas phase. Mobile (gold) and less mobile (copper) metals have been selected as model systems.

Project team of A2

Prof. Dr. Siegfried Blechert (TU Berlin)	Homogeneous catalysis, organic synthesis, natural product synthesis
Prof. Dr. Hans-Joachim Freund (FHI)	Spectroscopy and reactivity of metal clusters on oxide films
Prof. Dr. Gerard Meijer (FHI)	IR spectroscopy of metal and metal oxide clusters and their complexes
Prof. Dr. Joachim Sauer (HU Berlin)	Quantum mechanical studies of reaction mechanisms on surfaces and in clusters
Prof. Dr. Matthias Scheffler (FHI)	Electronic structure theory, density functional theory, first-principles statistical mechanics
Prof. Dr. Robert Schlögl (FHI)	In situ studies of model and real catalysts, synthesis and functional testing

A3: Hierarchically organised solid catalysts – Synthesis of advanced catalytic materials

Sub-Coordinator

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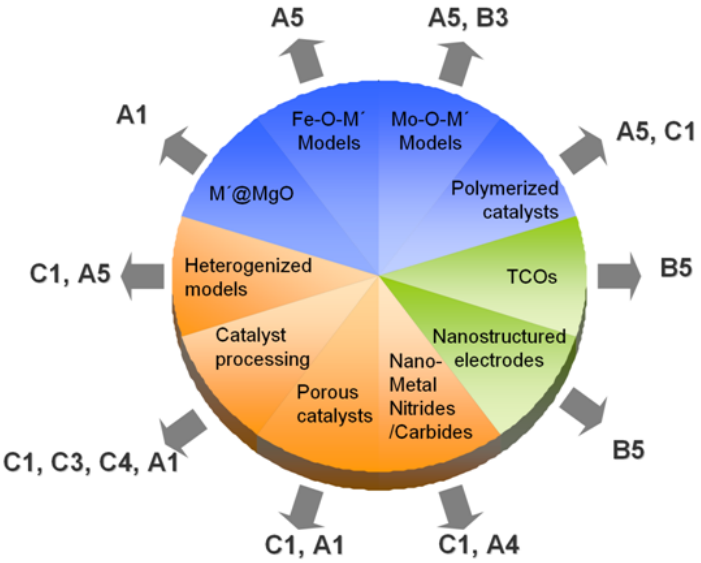
State of the art

The performance of a catalyst, i.e. its activity, selectivity and stability, is largely determined by its preparation. In this respect, synthesis is certainly one of the key steps and the most influential unit operation for the development of a successful catalytically active system. Three main targets are addressed:

- Advanced materials and molecular models for activation of small molecules (CH_4, O_2,…)
- Heterogenisation of homogeneous catalysts; functional/active supports
- Synthesis of controllable interfaces of materials/biology for bioelectronics

Research goals

This new research field is devoted to bridge the materials gaps by a knowledge-based molecular approach. Advanced catalytic materials will be developed for the fabrication of improved catalysts applied in the other research areas, taking advantage of synthetic methodologies in molecular, polymer and materials chemistry.



Another focus will be on low temperature synthesis to allow a precise control over chemical composition, surface chemistry, interfaces, pore structure, morphology, grain size etc.

The main objective is to provide synthetic and material solutions for emergent problems which arise throughout the course of the UniCat cooperation. Naturally, some materials are already in focus and will be synthesized or improved with the objective to control their structure on several length scales from the atomic scale (molecular models) to the nano- and microscale (nano/mesostructured materials) to the macroscale (supports).

Project team of A3

Prof. Dr. Matthias Driess (TU Berlin)	Metalorganic chemistry; Precursor synthesis; Molecular models for heterogeneous catalysts
Prof. Dr. Martin Lerch (TU Berlin)	Solid state chemistry
Prof. Dr. Michael Lehmann (TU Berlin)	Optical near field spectroscopy, TEM spectroscopy
Dr. Anna Fischer (TU Berlin)	Porous conductive materials, surface functionalization
Dr. Ralph Krähnert (TU Berlin)	Gas phase catalysis, porous catalysts
Prof. Dr. Robert Schlögl (FHI)	Heterogeneous catalysis, spectroscopy
Prof. Dr. Arne Thomas (TU Berlin)	Organic frameworks, functional materials

A4: Consecutive catalysis for fine chemical synthesis

Sub-Coordinator

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State of the art

Homogeneous catalysis provides indispensable tools for the preparation of a variety of important organic compounds, e.g. pharmaceutical intermediates, and is primarily target-oriented. Modern target-oriented synthesis aims at reducing the number of steps, increasing energy efficiency, lowering raw material consumption and avoiding by-products and waste. Nowadays, synthesis of organic fine chemicals and drugs produces more than three times as much waste as product (by weight), and the costs of recycling expensive metals in metal-containing catalysts can be high. Consequently, there is an enormous demand for the integration of several synthetic concepts in catalysis. Achieving very high specificity and selectivity in multi-step synthesis requires a synergetic approach that uses synthetic concepts from all types of catalysis: homogeneous, heterogeneous, and bio-catalysis.

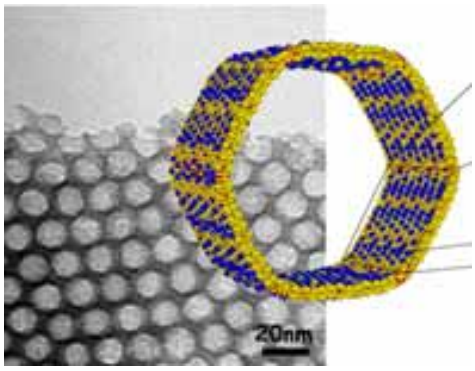

Fig. 4: Catalyst with a mesoporous structure.

Research goals

The implementation of these types of reactions is a key objective of the current project. “Tandem” reactions that involve the “sequential” utilisation of catalytic transformations with minimum workup, and concurrent “tandem” catalysis, in which multiple catalysts operate simultaneously, belong to the most efficient strategies in homogeneous catalysis for fine chemical synthesis. The complexity of “sequential” chemical transformations requires a knowledge-based strategy rather than the current trial-and-error approach. We make use of the generic understanding acquired in the other Research Fields of this Area. To systematically guide catalyst optimisation, mechanistic studies are performed including gas-phase studies of elementary steps, and dedicated quantum chemical calculations. Within the field of homogeneous catalysis we are searching for innovative catalysts suitable for new C–X coupling reactions (e.g. X = C, N, O), producing organic target molecules including enantiopure compounds that are of significant impact in the pharmaceutical industry.

Project team of A4

Prof. Dr. Markus Antonietti (MPI-KGF)	Mesostructured materials, sol gel chemistry, SAXS
Prof. Dr. Siegfried Blechert (TU Berlin)	Homogeneous catalysis, organic synthesis, natural product synthesis
Prof. Dr. Thomas Braun (HU Berlin)	Mechanistic studies, coordination chemistry
Prof. Dr. Matthias Driess (TU Berlin)	Molecular main group and organometallic chemistry, precursors, NMR
Prof. Dr. Rainer Haag (FU Berlin)	Hyperbranched and dendritic polymers, homogeneous catalysis
Prof. Dr. Karola Rück-Braun (TU Berlin)	Organic synthesis, catalysis, peptide chemistry
Prof. Dr. Helmut Schwarz (TU Berlin)	Gas-phase ion and physical organic chemistry
Prof. Dr. Roderich Süssmuth (TU Berlin)	Biocatalysis, enzymatics, natural products, antibiotics, structure elucidation

A5: Linking homogeneous and biocatalysis

Sub-Coordinator

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State of the art

Biological catalysts are known to activate small molecules and facilitate the manufacture of products of added value, usually at ambient temperature and pressure.
The aim is to establish a biomimetic approach which helps in obtaining reliable information about the electron transfer mechanisms in enzymatic catalysts, in collaboration with Research Area B. Conversely, the joint efforts to elucidate the reaction mechanisms of enzymes may enable the understanding of unifying principles that bridge the gap between homogeneous and biocatalysis. We want to develop a rational design of bioinspired compounds that aim to reach enzymatic efficiency, but at a lower level of complexity.

However, for a more detailed understanding of metalloenzymes on a molecular scale, it is first necessary to reduce the level of structural complexity. Low molecular-weight metal complexes can be suitable model compounds for analysing the structural and electronic parameters of active sites in metalloenzymes.

Depending on how far the exploration of the coordination active site(s) in the enzyme has advanced, biomimetic models can help to determine the nature of the active site or the effect of the coordination environment on its electronic structure, and hence its spectroscopic signatures. The information obtained for model compounds can then be used to determine the active site structures of intermediate states, on the basis of spectroscopic studies. Finally, such models may aid in the design of bio-inspired catalysts that aim to achieve performance similar to that of enzymes, albeit using a lower level of complexity.

Research goals

The goal of this Research Field is to establish a biomimetic approach to coordination compounds which helps to obtain reliable information about enzymatic reaction mechanisms in metalloenzymes, in collaboration with Research Area B. This aim is conceptually related to the in situ studies on the new Mo-based systems intended for methane activation and directly links the results in this Field and in Research Field A3.

The *biomimetic research* focuses on model systems for oxygenases such as the soluble methane monooxygenase (sMMO), as well as [FeFe]- and [FeNi]-hydrogenases. One important aspect concerns the redox cooperativity metal ←→ ligand or metal ←→ metal in conjunction with substrate and product binding (H_2, O_2, H_2O). Progress made in this field will certainly help to unravel the nature of active sites and to develop more effective artificial catalysts.

Project team of A5

Prof. Dr. Holger Dau (FU Berlin)	Metalloproteins, X-ray spectroscopy
Prof. Dr. Matthias Driess (TU Berlin)	Metal-organic chemistry, precursor synthesis, molecular models for heterogeneous catalysts
Prof. Dr. Bärbel Friedrich (HU Berlin)	Hydrogenases, biochemistry
Prof. Dr. Peter Hildebrandt (TU Berlin)	Redox proteins / vibrational spectroscopy
Prof. Dr. Christian Limberg (HU Berlin)	Oxygenation chemistry, metal oxo complexes, heterobimetallic compounds
Dr. Kallol Ray (HU Berlin)	Bioinorganic chemistry, synthesis of molecular models for sMMO

V. Research Area B: “Intelligent” natural and artificial enzymes

Coordinator

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A large variety of biological processes are controlled by enzymes which have been evolutionarily developed to catalyse complex chemical reactions with high efficiency and selectivity.

The research of Area B is guided by the concept of “creative learning from nature” and dedicated to the structural and functional analyses of complex natural enzymatic systems and the development of novel enzyme complexes for potential technological applications.

Selected catalysts, including three groups of metal-containing redox proteins and several light- and voltage-activated membrane-bound enzymes, are used as model systems to elucidate reaction mechanisms, structure-function relationships and dynamic processes on the molecular and modular level.

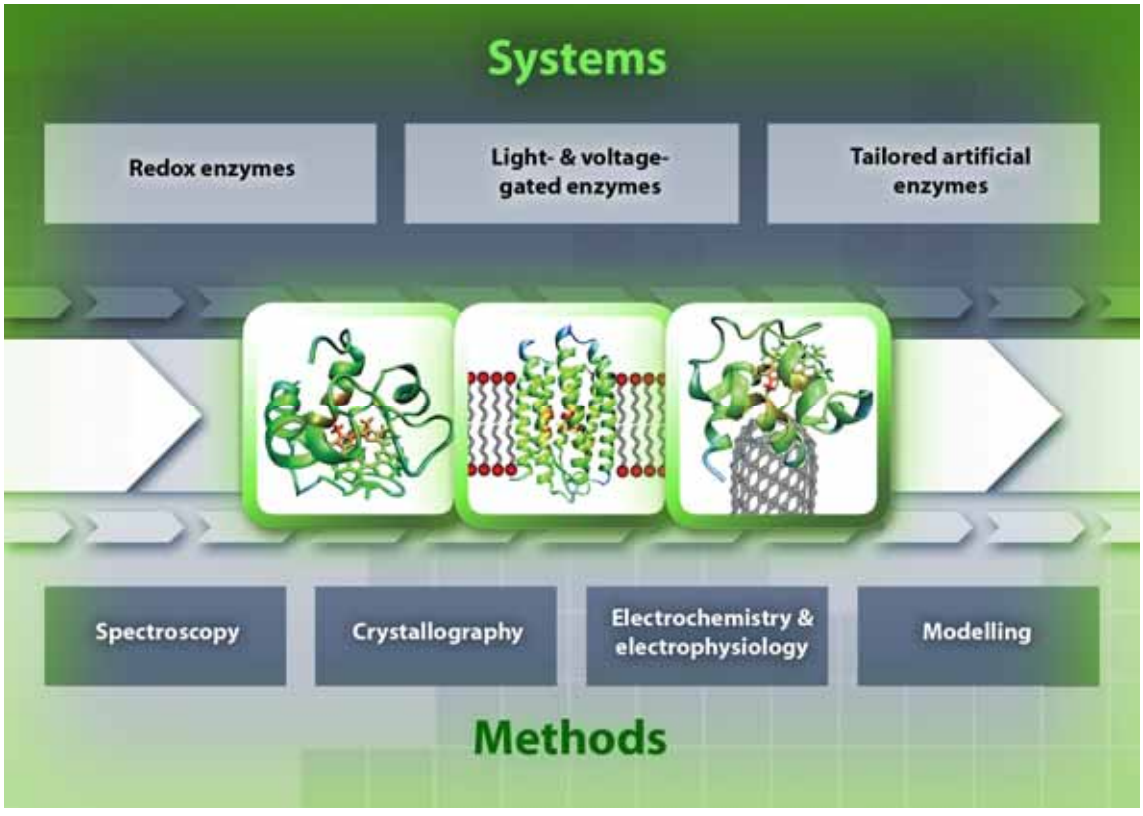

Fig. 5: Research structure in Area B indicating the hierarchical approach to complex catalytic systems.

Research goals

- Elucidation of the structures of complex enzymatic systems as well as the mechanisms and dynamics of biocatalytic processes by the concerted application of molecular genetics, synthetic molecular models (obtained in Area A), biochemical and structural techniques, and advanced spectroscopic and theoretical methods.
- Design and construction of artificial enzymes with tailored functions and novel sensor-enzyme complexes.

Research Fields of Area B

Research Area B is organised into five Research Fields.

B1: Photosynthetic water oxidation, light driven H2 production, hydrogenase-based biofuel cells

B2: Structure-function analysis of oxygen tolerant hydrogenases

B3: Cofactor insertion and functional investigations on complex molybdoenzymes

B4: Light- and voltage-gated enzymes

B5: Bioelectronic building blocks

B1: Photosynthetic water oxidation and bioelectronic devices

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Research Field B1 is divided into the two areas "Photosynthetic water oxidation" and "Bioelectronic devices for biofuel cells and light-driven hydrogen production".

Photosynthetic water oxidation

State of the art

Oxygenic photosynthesis is one of the central biocatalytic processes within the biosphere. Exploiting solar energy, plants and cyanobacteria convert water and carbon dioxide into the energy-rich carbohydrates that ultimately fuel life on earth. Not only energy but also precursors of biomass formation (H, C, O) become available by photosynthetic H_2O oxidation and CO_2 reduction, both of which are catalysed with high efficiency by protein-bound metal centres. As a by-product, atmospheric molecular oxygen is produced and continuously replenished. The enzymes involved belong to the most abundant proteins on earth.

The project focuses on water oxidation in association with the pentanuclear Mn4Ca complex bound to photosystem II (PSII), a membrane-intrinsic protein complex. Light-driven catalysis of H_2O oxidation is of fundamental importance but is still insufficiently understood. Studies in this field may provide insights into general principles of multi-step biological redox catalysis, which are also of potential technological relevance.

The aim of this interdisciplinary consortium is to develop, verify and apply new methods in order to make an essential contribution to elucidating the catalytic mechanism of the manganese cluster. The objectives of further improvements to the crystal structures are to fully understand light-driven water splitting by PSII and PSI.

Research goals

Specifically, the aims of the studies within this area are
(i) to develop methods for elucidating the electronic properties of the Mn_4Ca complex and
(ii) to track the transitions between intermediate states of the Mn_4Ca complex in the reaction cycles by time-resolved, structure-sensitive methods.

There is a specific focus on synchrotron radiation-based methods in order to study the electronic structure of synthetic model complexes and protein-bound metal centres. EPR spectroscopy will complement the XAS studies.

To probe the dynamics of oxygen formation, the Mn_4Ca complex of PSII will be driven through its catalytic cycle by a sequence of saturating nanosecond laser flashes and the various intermediates will be monitored by time-resolved K-alpha and K-beta fluorescence spectroscopy (ESRF) and RIXS experiments (resonant inelastic X-ray scattering). The potential of FTIR experiments for studying the oxidation-state changes of the Mn ions in PSII will be explored. Time-resolved crystallography would be the ultimate experiment to investigate protein movements associated with the catalysis of water oxidation on the PSII donor side. In a first step, laser-induced transitions in the catalytic cycle will be probed by time-resolved recombination-fluorescence measurements on PSII single crystals. Structure-based interpretations of the experiments will be conducted by molecular modelling and quantum chemical calculations for geometric and electronic structures.

Bioelectronic devices for biofuel cells and light-driven hydrogen production

Research on photosynthetic water oxidation and hydrogenases is linked by an exciting long-term prospect: the efficient coupling of both processes with the objective of producing hydrogen from light and water.

Research goals

O_2-tolerant [NiFe]-hydrogenases, which are investigated in Research Field B2, will be directly coupled with peripheral subunits of photosystem I (PSI) by different strategies. Proof of the concept of genetic fusion has recently been demonstrated and will be pursued by using different hydrogenases and PSI subunits for hybrid construction. In an alternative approach, conducted in collaboration with J. Golbeck (Penn State University), hydrogenases and PSI are electronically connected in vitro using chemically synthesised molecular wires.

The efficiency of these devices crucially depends on the tight electronic coupling of the components, which will be optimised by engineering the respective partners. Promising intermediates will be investigated by spectroscopic techniques available in UniCat.

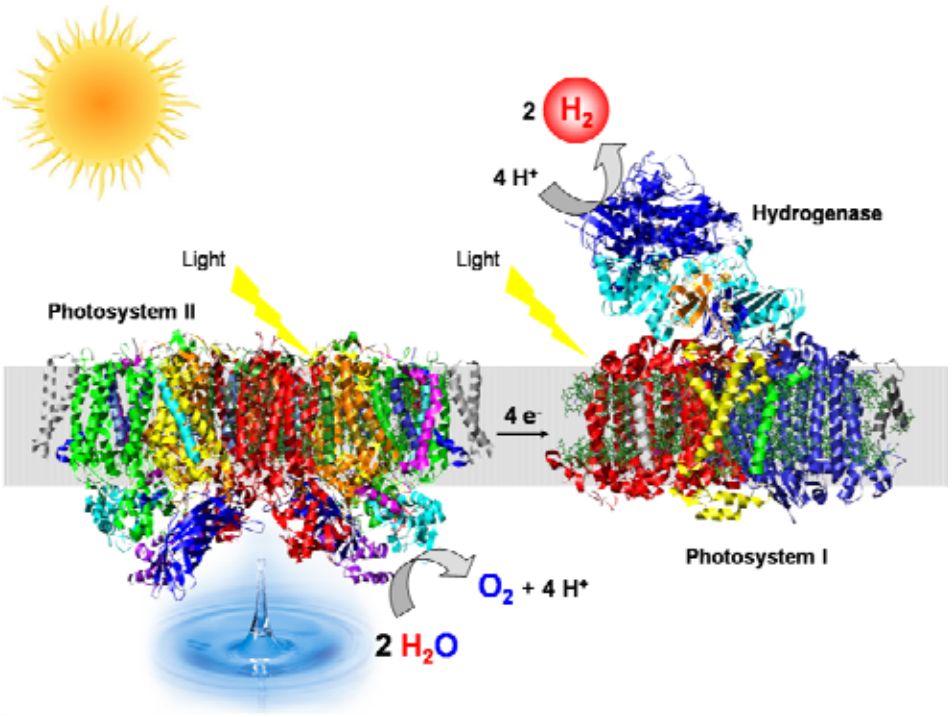

Fig. 6: Model of light-dependent hydrogen production from water by using cyanobacterial photosystems and oxygen-tolerant hydrogenase.

Another promising technological application, which is also based on the exceptional tolerance towards O_2 and CO of the [NiFe]-hydrogenases investigated in UniCat, is the development of enzyme-based fuel cells. In close collaboration with K. Vincent and F. Armstrong (University of Oxford), we have constructed a prototypical biological fuel cell that is equipped with enzyme-coated pyrolytic carbon strips instead of expensive platinum electrodes and does not require the membrane-based separation of the compartments. Our intention is to improve the power output and lifetime of the biofuel cell by isolating alternative O_2-tolerant hydrogenases from aerobic H_2-oxidizing bacteria. The studies will be done in close collaboration with Area C.

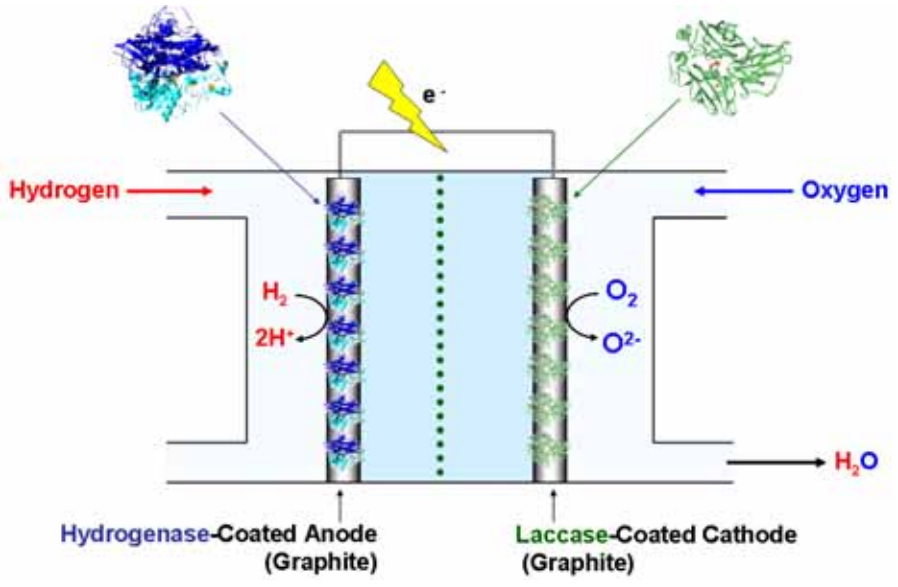

Fig. 7: Biofuel cell based on graphite electrodes covered with highly-specific biocatalysts.

Project team of B1

Prof. Dr. Holger Dau (FU Berlin)	XAS spectroscopy on catalysis at metal sites
Dr. Oliver Lenz (HU Berlin)	Application of hydrogenases
Prof. Dr. Athina Zouni (TU Berlin)	Biochemistry of photosystem I and II
Prof. Dr. Robert Bittl (FU Berlin)	EPR spectroscopy
Prof. Dr. Peter Hildebrandt (TU Berlin)	Vibrational spectroscopy
Prof. Dr. Martin Kaupp (TU Berlin)	Quantum chemical calculations
Prof. Dr. Maria Mroginski (TU Berlin)	Quantum chemical calculations
Prof. Dr. Ulla Wollenberger (Universität Potsdam)	Electrochemistry

B2: Structure-function analysis of oxygen-tolerant hydrogenases

Sub-Coordinator

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State of the art

The reversible cleavage of H_2 into protons and electrons is a prevalent metabolic process in microorganisms, catalysed by two major classes of enzymes, the [NiFe]- and the [FeFe] hydrogenases. This project centres on the elucidation of the redox chemistry catalysed by oxygen-tolerant [NiFe] hydrogenases. Their active site is composed of an intricate complex of transition metals, forming a heterodinuclear Ni-Fe cofactor that is coupled with intramolecular electron and proton pathways.

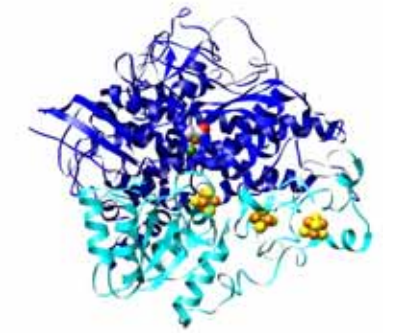

Fig. 8: Example of hydrogenase.

Crystal structures and advanced spectroscopic analyses combined with molecular and biochemical tools have paved the way to a basic understanding of the reaction mechanism. It is generally accepted that the nickel-iron centre in the [NiFe] hydrogenases provides a free coordination site for binding hydrogen. In the oxidised state of [NiFe] hydrogenases, this site is usually occupied by oxygen which has to be reductively removed prior to catalysis. The nature of the vacant site, of which subtle modifications may exist, provides one of the keys to understanding the catalytic properties of hydrogenases and offers potential targets for optimising the catalyst by genetic engineering.

Research goals

This Research Field focuses on the three [NiFe] hydrogenases in the ß-proteobacterium *Ralstonia eutropha*, which utilise the hydrogen-splitting reactions to perform different physiological functions, i.e. energy conservation, production of reducing equivalents and transcriptional regulation (hydrogen-sensing). Our major goal is to elucidate the molecular reaction mechanism of this group of [NiFe] hydrogenases, in particular the background for their oxygen tolerance and carbon monoxide insensitivity. Both features are crucial for the application of a hydrogen-activating catalyst. In general, oxygen may affect [NiFe] hydrogenases on three different levels, all of which are subjects of our research:

- posttranslational maturation
- catalytic activity
- regulation of gene transcription

Project team of B2

Prof. Dr. Bärbel Friedrich (HU Berlin)	Hydrogenases, physiology and biochemistry
Prof. Dr. Holger Dau (FU Berlin)	X-ray spectroscopy, EXAFS
Prof. Dr. Holger Dobbek (HU Berlin)	Cofactor-containing enzymes
Prof. Dr. Maria Andrea Mroginski (TU Berlin)	Theoretical approaches for spectroscopy
Prof. Dr. Peter Hildebrandt (TU Berlin)	Vibrational spectroscopy, photoreceptors
Prof. Dr. Robert Bittl (FU Berlin)	EPR spectroscopy
Prof. Dr. Martin Kaupp (TU Berlin)	Quantum chemical calculations
Dr. Oliver Lenz (HU Berlin)	Application of hydrogenases

B3: Cofactor insertion and functional investigations on complex molybdoenzymes

Sub-Coordinator

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The molybdenum cofactor (Moco) is the essential component of a group of redox enzymes which are of major importance in human metabolism. Sulfite oxidase (SO) is essential for humans and catalyses the oxidation of sulfite to sulfate, the last step in the degradation of sulfur-containing amino acids. Xanthine dehydrogenase (XDH) and aldehyde oxidase (AO) are complex metallo-flavoproteins that contain Moco, two [2Fe2S] clusters and flavin adenine dinucleotide (FAD) as catalytically acting units, and catalyse the oxidative hydroxylation of purines, pyrimidines, pterines, and aldehyde substrates using NAD+ or molecular oxygen as electron acceptors. The project centres on the mechanism of cofactor insertion and enzyme assembly and the spectroscopic characterisation of diverse molybdoenzymes.

The in-situ assembly of the holoprotein and the specificity of cofactor recognition and insertion is investigated by EXAFS studies and supported by studies on electrode surfaces via pre-immobilised cofactors and protein subunits. Structural changes upon cofactor insertion and protein assembly of different subunits and Moco-protein interactions are determined using EPR spectroscopy of spin-labelled proteins.

While the biochemical function of XDH is well established, the biochemical and physiological role of AO is still obscure. AO is believed to play an important role in the metabolism of drugs and xenobiotica. To elucidate the role of this enzyme in the mammalian organism and to define the specific physiopathological function, AO isoforms (mouse, human) were purified. The studies are paralleled by analysing similarities and differences of the active site structures and mechanisms of AO isozymes using various spectroscopic (IR, Resonance Raman, EPR, XAS) and electrochemical techniques. The studies are based on the recent progress achieved for the purification of mammalian AOs in larger amounts, and extending the scope of the research programme to include interdisciplinary approaches and novel methods developed within the Cluster will help lead to an understanding of the complex enzyme systems of molybdo-flavoenzymes in detail.

Project team of B3

Prof. Dr. Silke Leimkühler (Universität Potsdam)	Molybdoenzymes, biochemistry
Prof. Dr. Martin Kaupp (TU Berlin)	Quantum chemical calculations
Prof. Dr. Ulla Wollenberger (Universität Potsdam)	Electrochemistry, Biosensors
Prof. Dr. Thomas Risse (FU Berlin)	EPR Spectroscopy
Prof. Dr. Peter Hildebrandt (TU Berlin)	Vibrational spectroscopy, metalloenzymes
Prof. Dr. Robert Bittl (FU Berlin)	EPR Spectroscopy
Prof. Dr. Maria-Andrea Mroginski (TU Berlin)	Vibrational spectroscopy

B4: Light- and voltage-activated, membrane-anchored enzymes

Sub-Coordinator

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State of the art

Controlling the catalytic activity of enzymes is one of the central goals in biocatalysis. Nature has designed a large variety of control mechanisms of impressive robustness. These systems often comprise a combination of sensory and catalytic modules, the function of which relies on concerted conformational changes induced by the sensor. From an experimental point of view, systems using sensors for light, voltage, or redox potential are most attractive, because they represent well-defined and easy-to-control input signals.

In this Research Field, we will study enzyme complexes consisting of a catalytic and a light- or voltage-sensor module to gain insights into the structural and dynamic aspects involved in these regulation systems. In particular, we will use phosphatases and adenylate cyclases (ACs), which catalyse the production of phosphatidyl-inositol-biphosphate (PIP2 from PIP3) and cyclic-adenosyl monophosphate (cAMP) from ATP, respectively. The sensor modules of these systems are either membrane-associated blue-light receptors (BLUF), membrane-inserted green-light receptors (rhodopsins), or voltage sensors. Even though a substantial amount of information is already known for the individual modules, the properties of the coupled systems are largely unknown.

Our research will therefore concentrate on the spatial and dynamic properties of the conformational coupling process between sensor and catalyst, which will take advantage of the fact that enzymatic activity can most conveniently be triggered and synchronized by short light pulses or voltage steps. To understand the conformational fine-tuning of catalytically active sites, these investigations require close collaboration between researchers in spectroscopy, electrophysiology and structural biology.

Research goals

The goal of the project is the understanding of enzymes and catalysts that are modulated by light or membrane voltage, beginning from the sensing of the light absorption or voltage jump via early conformational changes in the chromophore domains, towards enzyme activation, adaptation and silencing. The five-year goal is to tailor new enzymes with specific chromophore or voltage sensor properties, and with catalytic activity that will be applicable to medical science and biotechnology (Optogenetic Technology).

Project team of B4

Prof. Dr. Peter Hegemann (HU Berlin)	Light-activated proteins, biochemistry and electrophysiology
Prof. Dr. Thomas Friedrich (TU Berlin)	voltage-activated enzymes, electrophysiology
Prof. Dr. Nico Ernsting (HU Berlin)	Ultra-fast spectroscopy on photoreceptors
Prof. Dr. Peter Saalfrank (Universität Potsdam)	Theory of photoreceptors
Prof. Dr. Robert Bittl (FU Berlin)	EPR Spectroscopy
Dr. Thomas Risse (FHI)	EPR Spectroscopy on surface-bond proteins
Prof. Dr. Peter Hildebrandt (TU Berlin)	Vibrational spectroscopy on photoreceptors
Prof. Dr. Maria Mroginski (TU Berlin)	Calculations of vibrational spectra

B5: Bioelectronic building blocks

Sub-Coordinator

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State of the art

Photosynthesis, respiration, redox signalling and metabolism include multi-step electron transfer processes. Basic principles of these biological redox systems are transferred to electrodes in bioelectronic devices based on bioelectrocatalysis at the electrode interface. Recently macroscopic surface modification has been substituted by tailor-made architectures such as carbon and semiconductor nanostructures, i.e. single-wall carbon nanotubes, doped carbon nanofibres, carbonaceous molecular sieves and nanoporous semiconductor films. Functionalistion of the conducting nanostructures (CNS) allows the binding of redox enzymes in order to exploit their highly specific catalytic properties for constructing bioelectronic sensors and biofuel cells.

Research goals

The research is focused on two enzyme families
(i) molybdenum enzymes and
(ii) hemeproteins.
The main goals are to elucidate fundamental aspects of electron transfer in nanostructured enzyme-CNS hybrids and to develop bioelectronic devices, such as bioelectronic sensors, enzyme nano reactors and biofuel cells.

The objectives of the project are twofold:

Fundamental aspects of electronic properties in CNS-enzyme hybrids will be investigated using spectroscopic and electrochemical techniques. This will also involve the development of novel spectroscopic techniques, such as time-resolved and near-field surface enhanced techniques, and the theoretical description of CNS-hybrid complexes. Bioelectronic building blocks on the basis of nanostructured architecture will be developed using redox proteins and enzymes and various nanostructured materials, such as nanoparticles, nanotubes and nano- and mesoporous carbon and semiconductor materials.

Initially within these projects small redox proteins will be explored, while later more complex heme and molybdenum enzymes will be used for bioelectrocatalysis, redox signalling and signal amplification in bioelectronic sensors for metabolites, nano-enzyme reactors for aromatic drugs and alkanol-based biofuel cells.

Project team of B5

Prof. Dr. Peter Hildebrandt (TU Berlin)	Redox enzymes, spectroelectrochemistry
Prof. Dr. Andreas Knorr (TU Berlin)	Nano- and near field optics, electron transfer
Prof. Dr. Janina Maultzsch (TU Berlin)	Optical near field spectroscopy
Prof. Dr. Frieder W. Scheller (Universität Potsdam)	Bioanalysis, biosensors, bioelectronics
Prof. Dr.Ulla Wollenberger (Universität Potsdam)	Electrochemistry, biosensors
Dr. Marga Lensen (TU Berlin)	Scanning probe microscopy
Dr. Anna Fischer (TU Berlin)	Design and synthesis of porous electrodes for enzyme immobilization

VI. Research Area C: Complex reaction engineering

Coordinator

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Novel catalysts are economically viable only if they can be used in industrial, large-scale processes. Scaling up from laboratory-scale experiments to industrial-scale processes is not simply enlargement of a new catalytic reaction; instead, many additional phenomena such as heat and mass transfer, have to be taken into account. These require sophisticated modelling, novel reactor geometries and concepts, and realistic cost estimates.

The Research Fields of Area C aim for the exploitation of chemically and biologically mediated catalytic processes. They provide bridges to both Research Areas A and B, thereby constituting the interface between fundamental research and industrial application.

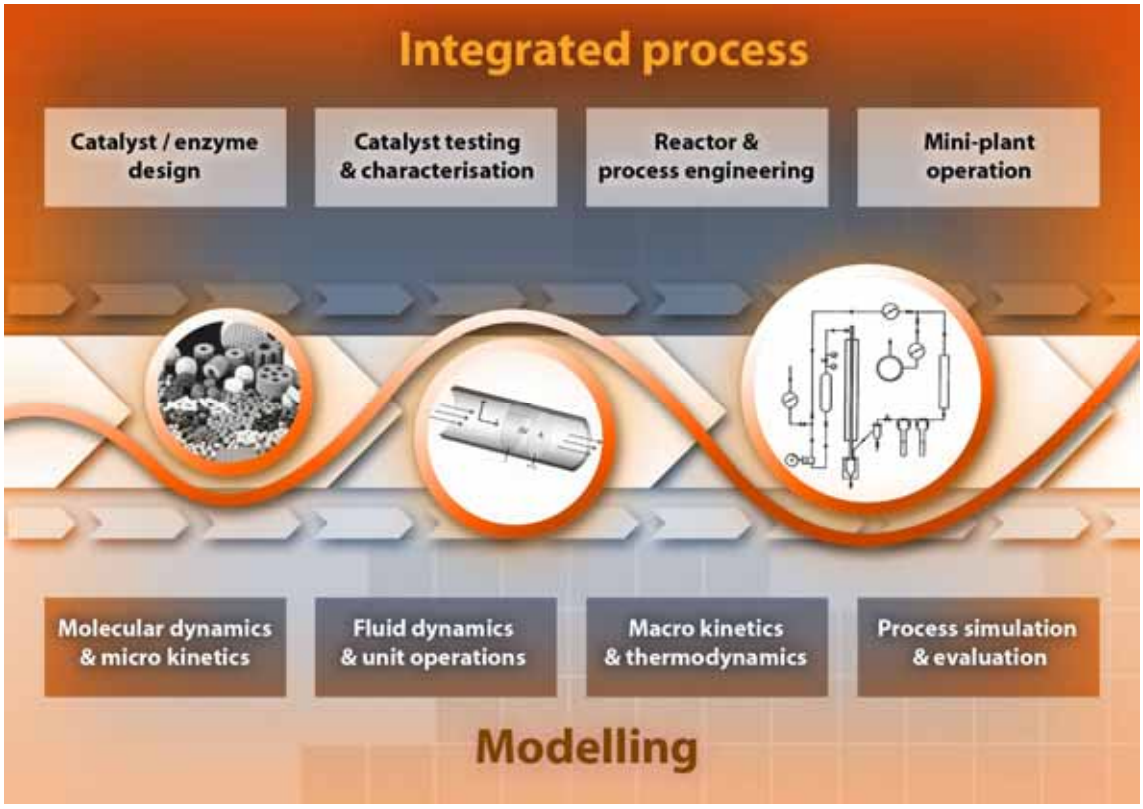

Fig. 9: Activities in Research Area C.

Research goals

The central objective of Research Area C is to transfer catalytic reactions to applications. This task requires the development of appropriate scaling-up procedures, as well as the design of new reactors and the optimisation of present reactor types. The operation of catalytic processes in miniplants requires a comprehensive simulation methodology to respond to the various underlying phenomena and reactions that occur on different time and length scales.

Hierarchical models are being developed that guide the evaluation and the management of integrated processes.

The challenge in biocatalysis is to exploit the potential of versatile and stable enzyme systems from the microbial primary and secondary metabolism for the synthesis of drugs and fine chemicals with high structural complexity.

Research Fields of Area C

Research Area C is organised into four Research Fields.

C1: Processing of solid catalysts

C2: Biocatalysis and process techniques

C3: Process simulation with hierarchic models

C4: Design of integrated catalytic processes

C1: Processing of solid catalysts

Sub-Coordinator

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State of the art

The activities of this Research Field are focused on the up-scaling, design, processing and characterisation of catalysts and support modules. The reactivity of a catalyst depends on the combination of materials and on a number of technical parameters, in particular the morphology of the support, the fluid dynamics, and the contact times of the reactants. Support materials can either be hard or soft matter and have to be customised for each catalytic system. Hard matter systems are chosen from the wide range of ceramic materials, while soft matter systems are based on self-assembling amphiphiles and functional polymeric structures such as polyelectrolytes, dendritic polymers and polymer gels.

Research goals

Promising catalytically active materials synthesised in Area A (e.g. catalysts based on molybdenum oxynitride; A3) are transformed into process-suitable catalysts. New catalysts are integrated into a more general reactor and process concept by close collaboration with C3 and C4 as well as by exchanging methods and materials with A1 and A2. For the immobilisation of homogeneous chemocatalysts and biocatalysts, methods based on new polymeric materials that enable efficient catalyst recovery by phase transfer or membrane separation processes are provided.

A particular challenge is the development of a durable porous support with a highly specific surface and long-life activity for the Li/MgO catalyst (midterm goal) by controlling the morphology of the support. Further enlargement of the catalytic surface would lead to smaller pores and, consequently, to undesirable mass-transfer limitations. This restriction of catalytic performance can be overcome by introducing a hierarchical pore structure. The methodology developed for the Li/MgO system will be adapted to other catalyst systems, e.g. Mo-O-N catalysts in the second five-year period of the CoE.

Project team of C1

Prof. Dr. Markus Antonietti (MPI-KGF)	Mesostructured materials, sol gel chemistry
Prof. Dr. Marion Ansorge-Schumacher (TU Berlin)	Biocatalysis, enzyme immobilisation
Prof. Dr. Rainer Haag (FU Berlin)	Hyperbranched and dendritic polymers, homogeneous catalysis
Dr. Constantin Czekelius (FU Berlin) *Associated member*	Hyperbranched and dendritic polymers, homogeneous catalysis
Prof. Dr. Regine von Klitzing (TU Berlin)	Thin films, microgel particles
Dr. Ralph Krähnert (TU Berlin)	Synthesis of porous materials, nanostructures, solid catalysts
Prof. Dr. Reinhard Schomäcker (TU Berlin)	Reaction kinetics, membrane reactors, reactor and process design
Prof. Dr. Helmut Schubert (TU Berlin)	Powder and materials technology, support and catalyst processing
Prof. Dr. Peter Strasser (TU Berlin)	High throughput testing, fuel cells, electrocatalysis

C2: Biocatalysis and process techniques

Sub-Coordinator

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State of the art

Peptides and polyketides from bacteria and fungi display various biological activities, e.g. antibacterial, antiviral, cytostatic etc., which are of interest for their application as drugs or for crop protection. These compounds are numerous and quite diverse in structure, as are their mechanisms of action, which makes them useful for the treatment of infections, immune diseases, cardiovascular dysfunction and cancer.

Peptide antibiotics are either produced by a ribosomal mechanism, e.g. nisin (food preservative), ziconotide (analgesic), or, more importantly, enzymatically by **n**on-**r**ibosomal **p**eptide **s**ynthetases (NRPS), e.g. vancomycin (antibiotic), bleomycin (cancer drug).

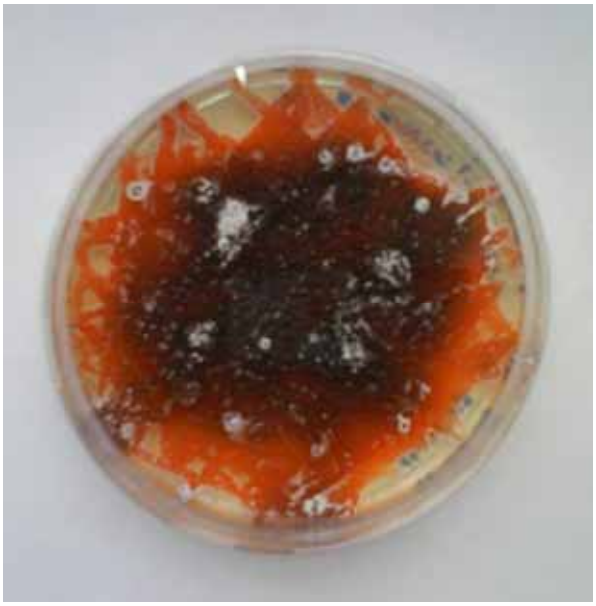

Fig. 10: Streptomyces: An example of an antibiotic-producing bacterium.

Polyketides like erythromycin (antibiotic) are assembled by polyketide synthases. A crucial point is the understanding of the biosynthesis of natural products on a genetic and protein level as a prerequisite for the optimisation of production or the structural alteration of the natural products. The NRPSs display a modular assembly of catalytic domains which, in a synthetic biology approach, enable the construction of new enzyme variants by combinatorial genetics and combinatorial biosynthesis.

From the exchange of these domains, the generation of new structural diversity in the metabolite is expected. Although much of the structural and functional basis of the mechanisms of these multifunctional enzymes has been elucidated, the engineering of available NRPSs and the design of new robust enzymes from existing modules is still in its infancy. Finally, concepts of the design of new proteins are investigated in a basic research approach. This includes the generation of artificial proteins and their modulation of enzymatic and spectroscopic properties.

Research goals

Sequencing and characterisation of novel NRPS gene clusters and their enzyme cascades involved in the biosynthesis of new natural products transfer of natural product biosynthesis from chemoenzymatics (in vitro) to combinatorial biosynthesis and mutasynthesis (in vivo) in heterologous hosts, e.g. E. coli establishing a library of modified NRPS modules and domains as a toolbox for the construction of new recombinant enzymes and the design of artificial enzymes and enzyme cascades for the generation of new structural diversity bioprofiling of new natural products for their bioactivity and druggability.

On request of the colleagues in the biotechnology department, the Research Field may be opened to other important enzyme classes. Engineers develop and support reactor concepts suitable for biocatalysts while providing their expertise to other subprojects. A particular focus is given to the support of the hydrogenase project (Research Field B2 and B4) which requires highly sophisticated fermentation conditions.

Project team of C2

Prof. Dr. Roderich Süssmuth (TU Berlin)	Combinatorial biosynthesis, synthetic biology and biological chemistry of natural products
PD Dr. Ullrich Keller (TU Berlin) *Associated member*	Biosynthetic enzymes of natural products
Prof. Dr. Rudibert King (TU Berlin)	Fermentation optimisation, modelling
Prof. Dr. Marion Ansorge-Schumacher (TU Berlin)	Hydrogenase biocatalysis
Prof. Dr. Peter Neubauer (TU Berlin)	Biotechnology of enzymes in industrial processes
Prof. Dr. Nediljko Budisa (TU Berlin)	Synthetic biology and molecular biotechnology of proteins
Prof. Dr. Siegfried Blechert (TU Berlin)	Substrate and natural product synthesis
Prof. Dr. Bärbel Friedrich (HU Berlin)	Hydrogenase characterisation

C3: Process simulation with hierarchic models

Sub-Coordinator

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State of the art

In chemical processes, many phenomena take place over different lengths and time scales. For each scale there are different numerical approaches and different programs. The different aspects of all of these scales must be integrated.

Research goals

A novel process simulator concept combining various methods at different levels of theory is being developed. On the one hand, the simulation starts with the results of ab initio quantum chemical methods for the prediction of molecular properties. The results will be used to describe the nanoscale behaviour up to the mesoscale processes. The transfer of these results to the mesoscale simulation with CFD and the macroscale simulation with the process simulator MOSAIC will be carried out. On the other hand, optimisation work on the macroscale has started considering uncertainties for the OCM processes Arco, UCC, Oxco, Suzuki and Turek-Schwittay, including gas purification processes, to determine the demands for catalyst development.

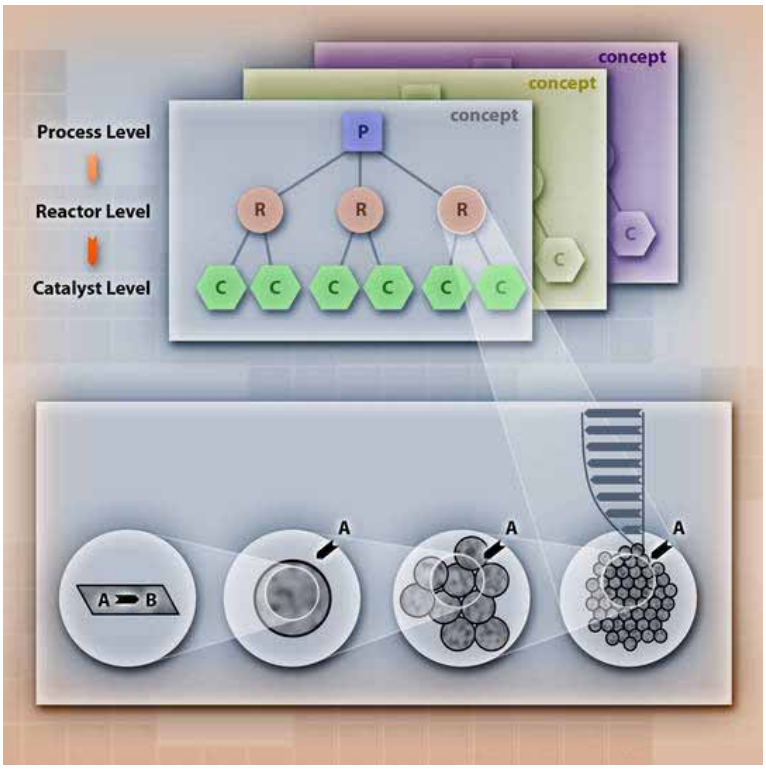

Fig. 11: Simulation of different process concepts with a hierarchical model for OCM.

A top-down approach and a classic bottom-up approach are being carried out in parallel. Demands for catalyst development will be defined in order to develop an economical process. Different reactor and downstreaming processes will be considered. Simulations on different levels will be integrated. Methods for experimental design and parameter identification for hierarchical models for OCM will be developed and applied to the OCM process.

This method will also be generalised for the biological process applications in Research Field C2.

Project team of C3

Prof. Dr. Matthias Kraume (TU Berlin)	CFD simulation
Prof. Dr. Günter Wozny (TU Berlin)	Process simulation, process optimisation
Dr. Raimund Horn (FHI, TU Berlin)	Micro-macro kinetic
Prof. Dr. Reinhard Schomäcker (TU Berlin)	Reaction engineering
Prof. Dr. Matthias Scheffler (FHI)	Kinetic Monte Carlo simulation (kMC), ab initio DFT
Prof. Dr. Joachim Sauer (HU Berlin)	Quantum mechanical studies of reaction mechanisms on surfaces and in clusters

C4: Design of integrated catalytic processes

Sub-Coordinator

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State of the art

As part of a close collaboration between chemical and process engineering, models for the individual reaction and process steps are used that allow for static and dynamic simulations of the overall process. On the basis of simulations, new process designs are derived and evaluated with respect to their potential for industrial implementation and economic efficiency. For this purpose, a new model-based cost-estimating procedure for the early design stage has been established. The most promising concepts are tested by constructing a mini-plant as an essential step towards further up-scaling for industrial applications. First assessments are done for processes using catalysts from Area A. The strategy will later be transferred to biocatalytic processes (collaboration with Area B and the professorships for Biocatalysis and Fermentation and Downstream Processing), in order to work towards a unifying concept for plant development in chemo- and biocatalysis.

Screening of catalysts and kinetic studies

A central task in this Research Field is the screening of catalysts with respect to activity, stability and selectivity. For catalysts with promising performance, more detailed kinetic studies are carried out. These investigations started with powder catalysts in order to obtain kinetic parameters for chemical reactions without any inference from heat or mass transfer limitations. For catalysts in homogeneous catalysis, the tests are carried out in collaboration with A4 and are expected to allow the design of appropriate reactors and process concepts. The integration of homogeneously catalysed reactions, catalyst recovery and product isolation within the same process unit offers the advantage of recycling without loss of activity or material.

Mini-plant

A mini-plant is under construction and is to be installed in order to provide a setting for checking new process concepts and assessing catalyst performance, start-up behaviour, recycling of feedstock and economy. Together with industrial partners, the potential for technological applications will be evaluated. Evaluation of new process concepts concerning safety assessments and toxicological impact are included from the beginning of process development. Safety assessment is of special importance since no concepts are available for this new type of integrated process operating at high temperatures with reaction mixtures in explodable/explosive regions. Besides acute safety aspects, long-term effects with potential toxicological relevance are investigated. The actual risks, however, depend on the bioavailability of the catalyst material; decisive factors are solubility, particle size, uptake and intracellular distribution. For example, almost nothing is known about the bioavailability and toxicity of

metal particles on nanoscale dimensions. Thus, depending on the materials to be selected in Areas A and B for further applications, the long-term toxicity, especially effects related to genotoxicity, are investigated in cell-culture systems. Based on previous investigations on relevant mechanisms of metal toxicity, special attention is paid to effects on protective processes involved in maintaining genomic stability, such as DNA repair processes, cell-cycle control and apoptosis. These toxicological investigations will promote the development of sustainable products, where potential risk factors are detected at an early stage of development and thus can be excluded to the greatest possible extent.

Project team of C4

Prof. Dr. Frank Behrendt (TU Berlin)	Reactive flows, porous reactive media, heterogeneous catalysis
Prof. Dr. Matthias Kraume (TU Berlin)	Transport phenomena, membrane bioreactors, reactor design
Prof. Dr. Reinhard Schomäcker (TU Berlin)	Reaction kinetics, membrane reactors, reactor and process design
Prof. Dr. Peter Strasser (TU Berlin)	High throughput testing, fuel cells, electrocatalysis
Prof. Dr. Günter Wozny (TU Berlin)	Process simulation, integrated processes, optimisation

Unifying Concepts in Catalysis

is the only natural science cluster of excellence in Berlin and Brandenburg. It is an interdisciplinary research network, whose central topic is catalysis, and within this framework future-related research topics concerning energy supply, bio-hydrogen and new chemical agents are addressed.

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