

Institute for Chemical Technology and Polymer Chemistry

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Prof. Dr. O. Deutschmann	Prof. Dr. JD. Grunwaldt	Prof. Dr. P. Théato	Prof. Dr. M. Wilhelm	Prof. Dr. C. Heske	Prof. Dr. F. Studt	Prof. Dr. R. Suntz	Prof. Deutschmann & Prof. Grunwaldt	
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Institute for Chemical Technology and Polymer Chemistry



The Institute for Chemical Technology and Polymer Chemistry comprises four divisions, two of them working in chemical technology and two in polymer chemistry. Additionally, three working groups in applied spectroscopy, applied computational catalysis and combustion diagnostic. The cooperative direction is formed by Prof. Deutschmann, Prof. Grunwaldt, Prof. Théato and Prof. Wilhelm.

The history of the institute starts in the year 1872, when the "Chemisch-Technisches Institut" was founded. The institute grew guickly and in 1921 was renamed to "Institut für Chemische Technik". Over 50 years later, in 1965, a new chair of Macromolecular Chemistry was established. The chair was assigned in 1967 to the newly founded "Polymer-Institute". The two institutes functioned independently until 2004 when the fusion under the name of "Institute for Chemical Technology and Polymer Chemistry" took place. Although the activity in the field of catalytic pollution control has a longer tradition at ITCP, the Exhaust Gas Centre Karlsruhe as a part of the institute was officially established only in 2010. In 2016 a new hard X-ray beamline for CATalysis and ACTinide/radionuclide research was successfully installed and commissioned at the KIT light source, which is jointly operated by research groups at the Institute for Chemical Technology and Polymer Chemistry, the Institute of Catalysis Research and Technology and the Institute for Nuclear Waste Disposal of KIT.

Over the years, the institute benefited from the contribution of notable professors, including Nobel laureates, which played a tremendous role in the development of the catalysis and polymer chemistry fields. Prof. Fritz Haber was awarded the 1918 Nobel Prize in Chemistry for his work on ammonia synthesis from its elements (the so-called Haber-Bosch process), a process discovered during his time at the former University of Karlsruhe (1894-1911) and further developed with Carl Bosch at BASF. Prof. Hermann Staudinger, who worked at the University of Karlsruhe between 1907 – 1912, received the Nobel Prize in Chemistry in 1953 for his contribution to the field of macromolecular chemistry.

Presently the institute has over 100 members and is located in several buildings at the Campus South, Campus North and Campus West of the KIT. The four main divisions in catalysis and polymer chemistry are:

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Applied Computational Catalysis Prof. Dr. F. Studt	30
Combustion Diagnostic Prof. Dr. R. Suntz	32
Exhaust Gas Center Karlsruhe Prof. Deutschmann, Prof. Grunwaldt	36

The main research topics in chemical technology are indepth understanding, simulation and optimization of catalysts for different reactions. In polymer science, novel methods for polymer synthesis for modern materials as well as their characterization are the research topics.

Climate and environmentally friendly novel chemical technologies

Chemical Technology

Prof. Dr. O. Deutschmann

The Deutschmann group works on the development of climate and environmentally friendly chemical technologies. The research activities include carbon-free chemical energy carriers, emission control, fuel and electrolysis cells, reaction engineering, heterogeneous catalysis, material synthesis, and multiphase flows. The methods used range from laser spectroscopy and kinetic measurements over digitalization and software development to numerical simulation and optimization of technical reactors.

There is a tremendous need for novel chemical technologies that are climate and environmentally friendly yet also economical viable. In close collaboration with many industrial and academic partners worldwide, we are guided by a holistic view on nature, technology, economy, and welfare.

Therefore, we develop and optimise sustainable technologies for industrial processes, clean transportation, and reliable energy supply. Our current research focuses on the reduction of emissions of greenhouse gases as well as local pollutants. We use advanced experimental and theoretical methods to study the kinetics of homogenous and heterogenous thermal and electro-chemical reactions and their interactions with mass and heat transport in chemical reactors, emission control devices, fuel and electrolysis cells. A special emphasis is given to the development of mathematical models and computer codes for the numerical simulation of technical processes.

Investigation of catalytic coated monoliths for gas conversion.

Our students are enrolled in both the Department of Chemistry and Biosciences and the Department of Chemical and Process Engineering. In catalysis research, we are closely linked to the Institute of Catalysis Research and Technology and the Grunwaldt group, with whom we collaborate on emission control in the Exhaust Gas Center Karlsruhe and the CRC 1441 TrackAct. In the ENERMAT (energetic materials) laboratory, we study fuel and electrolysis cells together with EIFER, the European Institute for Energy Research. Within the CRC/TRR150 and the initiative Clean Circles, we closely collaborate with TU Darmstadt on reactive flows and carbon-free chemical energy carriers, respectively.

Reduction of green-house gas emissions

We are engaged in the reduction of emissions of the greenhouse gases CO₂, CH₄, and N₂O from processes in chemical industry, traffic, transportation, and energy-intensive industries as well as the development and optimization of novel and existing chemical technologies. Hydrogen as climate-friendly energy carrier is of special importance. The production of hydrogen and the capture of carbon are investigated by methane pyrolysis in moving bed reactors. Methane coupling by high-temperature catalysis is studied as an alternative process for ethylene and acetylene production directly from natural gas.

Carbon-free chemical energy carriers

Carbon-free energy carriers are studied for storing renewable electrical energy in a circular economy. Here we focus on ammonia, metal borohydride such as KBH₄, and reactive metals such as iron and aluminium. For instance, sun and wind energy can be chemically stored by reducing iron oxides with electrolysis hydrogen in regions of inexpensive renewable energy. The time-flexible energy release through iron oxidation then takes place in regions with



▲ Fig. 2 Proposed energy cycle for iron as recyclable metal fuel.

The utilization of CO₂ is studied in catalytic dry reforming of natural gas for the production synthesis gas (Fig. 1) and for cleaning the off-gases of the steel industry [1] as well as for methanol production and methanation using green hydrogen. Even internal combustion engines are applied to serve as CO₂ reformer using this greenhouse gas instead of producing it.



▲ Fig. 1 Reforming of natural gas using CO₂; A. Giehr et al., Ind. Eng. Chem. Res. 59 (2020) 18790.

high energy demand (Fig. 2). Such seasonal, non-toxic, long-term energy carriers provide energy capacity that is no longer available due to the shutdown of nuclear and coal-fired power plants. They additionally compensate fluctuations in domestic renewable electricity, use existing large-scale infrastructures, and secure a political-independent energy supply.

Emission control

The reduction of emissions of local pollutants (NO_x, CO, aldehydes, particulate matter, etc.) arising from combustion engines, industrial plants, and residential sources is achieved by advanced exhaust-gas after-treatment systems. A special focus is given to the reduction of NO_x emissions by selective catalytic reduction (SCR) [2], the elimination of toxic pollutants such as formaldehyde and HCN [3], and better reaction engineering strategies for

the removal of the green-house gases such as CH₄ and N₂O [4]. Advanced capillary techniques and laser spectroscopy such as PLIF supports the understanding of the catalytic devices for cleaning exhaust gases. Slip of the greenhouse gases methane and N₂O from modern engines fueled by natural gas and hydrogen (Fig. 3), resp., are reduced by the application of advanced catalysts and optimised operating conditions.



▲ Fig. 3 H₂ as fuel for clean combustion engines; P. Lott et al., Chem. Ing. Techn. 94 (2022) 217.

High-temperature fuel and electrolysis cells

Our current research projects in this area focus on electrochemical devices, operated at high temperatures, for conversion of chemical into electrical energy and vice versa to serve the needs of a climate-friendly energy supply. We study solid-oxide cells and stacks as modern tools for hydrogen production by H₂O electrolysis, the co-electrolysis of H₂O and CO₂ for the production of synthesis gas [5], as well as the electro-chemically supported synthesis of chemical energy carriers such as ammonia. Solid-oxide fuel cells (SOFC) are applied for providing electrical power, which is also needed for mobility and transportation (Fig. 4).



▲ Fig. 4 SOFC for mobility and transport; L. Wehrle et al., ACS Environ. Au 2 (2022) 42.

Digitalization

A very strong focus of the group is on the development of models and computer codes for the description and numerical simulation in reaction engineering and catalysis. In academia and industry world-wide, software packages such as DETCHEM and CaRMeN (www.detchem.de), arising from our group, are used for the development of novel and optimization of operating conditions of existing processes in sectors such as chemistry, transport, and energy. Our current focus is on developing digital twins, efficient research data management, and artificial intelligence tools.

Selected papers

- 🗈 S. Angeli, S. Gossler, S. Lichtenberg, G. Kass, A. Agrawal, M. Valerius, K. P. Kinzel, O. Deutschmann; Reduction of CO₂ emission from off-gases of steel industry by dry reforming of methane; Angew. Chem. Int. Ed. 60 (2021) 11852.
- Progress in Energy and Combustion Science 87 (2021) 100949.
- removal by ammonia SCR in the exhaust of lean-burn natural gas engines; Angew. Chem. Int. Ed. 59 (2020) 14423-14428.
- tertreatment System; Emiss. Control Sci. Technol. 7 (2021) 1.
- 🖻 A. Banerjee, Y. Wang, J. Diercks, O. Deutschmann; Hierarchical Modeling of Solid Oxide Cells and Stacks producing Syngas via H₂O/CO₂ Co-electrolysis for Industrial Applications; Appl. Energy 230 (2018) 996.



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2 M. Börnhorst, O. Deutschmann; Advances and challenges of ammonia delivery by urea-water sprays in SCR systems;

3 D. Zengel, P. Koch, B. Torkashvand, J.-D. Grunwaldt, M. Casapu, O. Deutschmann; Emission of toxic HCN during NOx

P. Lott, O. Deutschmann; Lean-Burn Natural Gas Engines: Challenges and Concepts for an Efficient Exhaust Gas Af-

Composition of the team

Head: Prof. Olaf Deutschmann Deputy: Prof. Rainer Suntz Group leaders: Dr. Sofia Angeli Dr. Hendrik Goßler Dr. Thomas Häber Dr. Matthias Hettel Dr. Patrick Lott Dr. Steffen Tischer Dr. Martin Wörner

Olaf Deutschmann studied physics and chemistry in Magdeburg, Berlin, and Heidelberg. In 1996, he obtained his doctoral degree (chemistry) from Heidelberg University supervised by Jürgen Warnatz. As a Postdoc he worked on high-temperature catalysis at the University of Minnesota with Lanny D. Schmidt and at the Los Alamos National Laboratory. After receiving the venia legendi in Physical Chemistry from Heidelberg University, he joined the Faculty of the Department of Chemistry at the University of Karlsruhe, now KIT, in 2003.

Olaf Deutschmann organises the bi-annual MODE-GAT conference series and serves as member of several editorial and company advisory boards. He is recipient of the DECHEMA Award, the Hermann-Oberth-Medal, and Fellow of The Combustion Institute. The software packages DETCHEM and CaRMeN were developed under his guidance. He is also member of the board of directors at the Institute of Catalysis Research and Technology.







Schematic illustration of the electro- and thermo-catalytic chemistry as well as the coupled transport phenomena occurring during operation of a direct internal reforming solid oxide fuel cell (SOFC). The description of CH₄ reforming on the Nibased fuel electrode requires an elementary step mechanism. Collaboration with R.J. Kee, CSM, USA.

section of a selective catalytic reduction (SCR) system for removal of NO_x. Injected urea-water solution can impact on pipe walls, which can lead to liquid film and solid deposit formation. Test bench for the kinetic investigations of catalytic materials as

▲ Scheme of relevant physical and chemical processes in the mixing

function of pressure, temperature, gas-phase composition and flow rate. Included are the gas feeding section, the reactor containing the catalyst and devices for gas analysis such as FTIR, GC and MS.



 $H_2(g) + (Ni) + (Ni) \rightleftharpoons H(Ni) + H(Ni)$ $O_2(g) + (Ni) + (Ni) \implies O(Ni) + O(Ni)$ $CH_4(g) + (Ni) \rightleftharpoons CH_4(Ni)$ $H_2O(g) + (Ni) \rightleftharpoons H_2O(Ni)$ $CO_2(g) + (Ni) \rightleftharpoons CO_2(Ni)$ $CO(g) + (Ni) \rightleftharpoons CO(Ni)$ $O(Ni) + H(Ni) \rightleftharpoons OH(Ni) + (Ni)$ $OH(Ni) + H(Ni) \Longrightarrow H_2O(Ni) + (Ni)$ $OH(Ni) + OH(Ni) \rightleftharpoons O(Ni) + H_2O(Ni)$ $O(Ni) + C(Ni) \rightleftharpoons CO(Ni) + (Ni)$ $O(Ni) + CO(Ni) \Longrightarrow CO_2(Ni) + (Ni)$ $HCO(Ni) + (Ni) \Leftrightarrow CO(Ni) + H(Ni)$ $HCO(Ni) + (Ni) \implies O(Ni) + CH(Ni)$ $CH_4(Ni) + (Ni) \rightleftharpoons CH_3(Ni) + H(Ni)$ $CH_2(Ni) + (Ni) \rightleftharpoons CH_2(Ni) + H(Ni)$ $CH_2(Ni) + (Ni) \rightleftharpoons CH(Ni) + H(Ni)$ Fue $CH(Ni) + (Ni) \rightleftharpoons C(Ni) + H(Ni)$ $O(Ni) + CH_4(Ni) \rightleftharpoons CH_3(Ni) + OH(Ni)$ $O(Ni) + CH_3(Ni) \rightleftharpoons CH_2(Ni) + OH(Ni)$ $O(Ni) + CH_2(Ni) \Longrightarrow CH(Ni) + OH(Ni)$ $O(Ni) + CH(Ni) \rightleftharpoons C(Ni) + OH(Ni)$



Modeling of a fixed-bed catalytic reactor using detailed surface chemistry for catalytic partial oxidation of methane for the production of hydrogen.





While the interaction of bubbles with solid internals is ubiquitous in bubble column reactors, its details are not well understood. Coordinated experiments and numerical simulations show that as a bubble rises against a cylinder a separating liquid film is formed, which eliminates the effects of wettability on the cutting process.





Typical time and length scales in a bundled tube reactor for the conversion of chemicals using catalytic particles. The system includes macroscale processes as concevtive mass transfer down to molecular processes as reaction on the inner surface of the porous catalyst structure.

The reduction of green-house gas emissions requires the adaptation of many processes in chemical and steel industry, in the mobility and transport sector as well as energy supply. We develop new and optimize existing technologies for all processes shown. Carbon-free energy carriers play a crucial role for climate-friendly industry and society.

From the nanoparticle to the catalytic process: design, reaction and understanding with advanced spectroscopic and microscopic methods

Chemical Technology and Catalysis Prof. Dr. Jan-Dierk Grunwaldt



The Grunwaldt group is engaged in the development of new heterogeneous catalysts for sustainable production of chemicals, clean air and energy conversion technologies. The synthesis of new materials is based on novel preparation methods, knowledgebased design and in-depth understanding. Of particular focus are in situ and operando spectroscopy, including experiments at the XAS beamline CAT-ACT of KIT and international synchrotron radiation sources.

More than 90% of all chemical processes rely on catalysis. As depicted in Fig. 1, the group conducts research in the fields of energy conversion, fine chemistry, and environmental catalysis including emission control. For a targeted design of catalysts, an iterative strategy based on structure-performance relations is applied. The catalytic activity as well as structural, physical, and electronic properties are analysed using advanced characterization tools to optimise the synthesis methods. For the design of new materials, both conventional and novel preparation techniques such as flame spray pyrolysis, hydrothermal synthesis, and colloidal methods including robot-controlled synthesis are applied and further developed with international collaboration partners.

 Installation of an operando cell for a combined catalytic and X-ray absorption spectroscopy (XAS) experiment at a Synchrotron Radiation Facility.



▲ Fig. 1 Our research focuses on the design, testing and advanced characterization of new catalysts within biomass conversion, fine chemistry and emission control catalysis.

Energy-related catalysis

The demand for materials and processes to address the efficient long-term storage of electrical energy is steadily increasing, as fluctuating wind and solar energies are progressively incorporated into the energy system. To ensure a steady supply, the produced energy can be stored in the form of chemical energy carriers. In this respect, the Grunwaldt group focuses on both the electrochemical reactions for generating H₂, as well as its combination with CO/CO₂ to produce energy-dense molecules, such as methanol, higher alcohols and methane. The catalytic materials are investigated for dynamic structural changes under realistic reaction conditions and variations of the feed composition, which mimic the intermittent nature of renewable energy (Fig. 2) [1]. This research is the starting point of the DFG priority program SPP2080 and several additional funding initiatives of BMBF.



▲ Fig. 2 Schematic of using renewable resources for the production of sustainable energy, fuels and chemicals.

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ler to shaped pic reactors do studies ure reactions O ₂ soved profiles		Operandα XAS, XES DRIFTS, ATR-IR, Raman- spectroscopy Synchrotron methods: XES, XES, HERFD-XAS, spatially and time- resolved Digitalization				
nthesis Gas, Methanol Synthesis,						

Biomass conversion

The production of commodity chemicals from biomass is an important alternative to the power-to-chemicals strategy and addresses issues such as sustainability and carbon footprint. For this purpose, bio-derived platform chemicals are catalytically transformed (via hydrogenation, hydrodeoxygenation, oxidation, Diels-Alder reaction, etc.) into valuable chemicals, solvents, and polymer building blocks [2]. This includes the upgrading of bio-oil. The aim is to develop novel catalysts and sustainable processes that operate in an efficient way and under "green chemistry" principles (Fig. 2).

Environmental catalysis and clean air

Reducing air pollution represents one of the most essential topics in our society, strongly supported by progressively tighter emission regulations in EU countries and all around the world. We focus on both mobile and stationary sources, including indoor air guality. To develop highly efficient and durable after-treatment systems for exhaust gases, comprehensive characterization and testing are conducted. We apply this strategy for evaluating model or commercial diesel oxidation catalysts, three-way catalysts, methane oxidation, soot oxidation, and catalysts for removal of NO_x or volatile organic compounds. As part of the CRC TrackAct initiative, we aim at predicting, manipulating and designing more efficient catalysts for emission control by systematically understanding the transient behaviour and the nature of active sites at different length scales and complexity levels for mono- and bimetallic noble metal-based catalysts. Recently we were

able to uncover that the formation of small Pt clusters is a necessary step for the CO oxidation onset on Pt/CeO₂ single site catalysts (Fig. 3) [3].





In situ / operando characterization of heterogeneous catalysts

Modern characterization techniques, preferentially performed under realistic reaction conditions with simultaneous analysis of catalytic performance (i.e. "operando"), play a pivotal role in our research. In situ cells have been designed for the study of gas and liquid-phase reactions at high temperatures (up to 1100 °C) and high pressure (up to 150 bar) by means of X-ray absorption, UV-vis, infrared and Raman spectroscopy, as well as X-ray diffraction. Combined setups for complementary techniques simultaneously provide information about the crystalline,

X-ray amorphous structure, and surface species. Combining spectroscopic and kinetic data sheds light onto catalytic reaction mechanisms, even at temperatures up to 1000 °C [4]]. Group members frequently perform X-ray absorption spectroscopy and microscopy experiments at KIT, PETRA III, ESRF, SLS, SOLEIL and many other synchrotron light sources. A beamline dedicated to actinide and catalysis research is now running at the KIT synchrotron (CAT-ACT beamline) in close collaboration with scientists at the institutes INE and IBPT.

X-ray tomography

The micro- and nanofocused X-ray beams generated at modern synchrotron radiation sources have recently started to offer intriguing new opportunities for catalyst characterization by means of X-ray microscopy (XRM). This collective technique allows the many interactions of X-rays with matter to be exploited for the generation of images, e.g. diffraction, fluorescence, absorption or phase contrast (Fig. 4). By further extending these imaging techniques for the purpose of tomography, it is possible to generate 3D spatially-resolved volumetric maps of catalyst particles, beds, pellets, and reactors. Activities in XRM are specifically focused on: (i) design of in situ cells for acquiring 2D and 3D information under operating conditions, (ii) spatially resolved information, such as porosity, active site distribution, and investigation of deactivation processes, e.g., sintering or phase transformation, even operando [5], (iii) application of correlative or combined techniques, together with electron microscopy, XAS,

and other spectroscopic techniques. Where possible, catalyst characterization is performed in situ / operando, with high spatial resolution, and on realistic rather than model samples.



▲ Fig. 4 Imaging catalysts at hierarchical length scales.

Selected papers

- 🗈 K. F. Kalz, R. Kraehnert, M. Dvoyashkin, R. Dittmeyer, R. Gläser, U. Krewer, K. Reuter, J.-D. Grunwaldt; Future challenges in heterogeneous catalysis: understanding catalysts under dynamic reaction conditions; ChemCatChem 9 (2017) 17.
- 2 O. R. Schade, F. Stein, S. Reichenberger, A. Gaur, E. Saraci, S. Barcikowski, J.-D. Grunwaldt; Selective Aerobic Oxidation of 5-(Hydroxymethyl) furfural over Heterogeneous Silver-Gold Nanoparticle Catalysts; Adv. Synth. Catal. 362 (2020)
- F. Maurer, J. Jelic, J. Wang, A. Gänzler, P. Dolcet, C. Wöll, Y. Wang, F. Studt, M. Casapu, J.-D. Grunwaldt; Tracking the formation, fate and consequence for catalytic activity of Pt single sites on CeO₂; Nature Catal. 3 (2020) 824.
- 🕙 D. Eggart, A. Zimina, G. Cavusoglu, M. Casapu, D. E. Doronkin, K. A. Lomachenko, J.-D. Grunwaldt; Versatile and high temperature spectroscopic cell for operando fluorescence and transmission x-ray absorption spectroscopic studies of heterogeneous catalysts; Rev. Sci. Instrum. 92 (2021) 023106.
- ical gradients in automotive Cu-SSZ-13 catalysts for NO_x removal revealed by operando X-ray spectrotomography; Nature Catal. 4 (2021) 46.



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🖻 J. Becher, D. F. Sanchez, D. E. Doronkin, D. Zengel, D. M. Meira, S. Pascarelli, J.-D. Grunwaldt, T. L. Sheppard; Chem-

Composition of the team

Head: Prof. Jan-Dierk Grunwaldt Group leaders: Dr. Maria Casapu Dr. Dmitry Doronkin Dr. Erisa Saraçi Dr. Thomas Sheppard Dr. Anna Zimina

Jan-Dierk Grunwaldt studied chemistry at the University of Hamburg and at Newcastle upon Tyne (GB). After completing his Ph.D. at ETH Zurich in 1998, he joined the Danish catalysis and chemical engineering company Haldor Topsøe A/S as project leader in synchrotron radiation and applied catalysis, before moving back to academia in 2001 at ETH Zurich. In 2008, he became a full Professor of Chemical Engineering and Catalysis at the Technical University of Denmark (DTU). In 2010, he was appointed Professor of Chemical Technology and Catalysis at the Karlsruhe Institute of Technology.

He is a member of the board of directors at the Institute of Catalysis Research and Technology, Adjunct Professor at DTU, and has been a visiting professor at the University of Padua in 2022. Jan-Dierk Grunwaldt is the author of more than 400 publications, and has received several awards, for example, the Jochen Block Award (DECHEMA) and the Dale Sayers Award (International XAFS Society). He serves in various boards at the German, European and International Catalysis Societies (GeCatS, EFCATS, IACS, ICEC), at synchrotron radiation sources (PETRA III, BESSY, ESRF, SLS), International X-ray absorption spectroscopy (IXS) and the German Committee for Research with Synchrotron Radiation (KFS).



Chemical Technology and Catalysis Prof. Dr. Jan-Dierk Grunwaldt





▼ Chemical imaging of catalyst composition by hard X-ray tomography at PETRA III Synchrotron Radiation Facility (Hamburg, Germany).











▲ Catalyst testing benches at the Exhaust Gas Center Karlsruhe.

Pt/CeO₂ catalyst at work: ETEM images during redispersion of Pt particles in oxygen containing atmosphere.

▼ Enhancing the performance of emission control catalysts by *in situ* tuning the noble metal structure.

▲ CAT beamline at the KIT light source.

 Two-nozzle flame spray synthesis of nanoscaled catalysts.

High pressure cell for in situ/operando spectroscopic investigations of catalysts.



▼ Preparation of an operando X-ray absorption spectroscopy experiment at the European Synchrotron Radiation Facility (ESRF, Grenoble, France).

Polymer synthesis: novel materials development

Preparative Macromolecular Chemistry Prof. Dr. Patrick Théato

Current research in the team of Prof. Théato at the Karlsruhe Institute of Technology is situated at the interface of organic, polymer, and materials science. We focus on functional polymer synthesis, surface chemistry, nanomaterials, and materials science, by utilizing numerous modern polymerization techniques in combination with novel and efficient post-polymerization modification techniques. Further, with the development of new powerful synthetic tools, we are extending and pushing the current limits of polymer chemistry. The advanced development of precision synthesis of macromolecules utilizing novel chemistries leads to defined polymer materials. This opens the door to multiple applications, such as hydrogel actuators and energy storage materials.

Novel synthetic approaches

For several years, the group has developed new synthetic approaches for the synthesis of polymers. We usually transfer knowledge from organic small molecular chemistry to be adopted in the synthesis of polymers. Examples range from classical chemical ligation chemistries to multicomponent reactions. Further, continuous flow synthesis to control polymerizations and beyond is developed.

Post-polymerization modification

The group has a long lasting and renowned expertise in developing new synthetic approaches for the post-polymerization modification of reactive precursor polymers. In particular, the group pioneered the utilization of activated esters on the basis of pentafluorophenyl esters, which proved to be robust and efficient tools for the synthesis of amide-based polymers. Novel developments deal with the elimination of functional groups, essentially leading the way for a novel synthetic route toward polyethylene.

Smart polymer materials

Derived from reactive polymers, the group has a renowned expertise in the preparation of smart polymer materials. In particular the combination of different stimuli has been the focus of our research. Stimuli of interest include temperature, light, redox, pH, CO₂, or small moelcules such as glucose.

Hydrogels & actuators

Advancing smart polymers into cross-linked material results in smart hydrogels featuring a reversible actuation, which have received particular attention and have been further developed into sensors, actuators, or artificial muscles. Such functional three-dimensional polymer networks are prepared with the capability of shape deformations showing a volume phase transition after being triggered by external physicochemical stimuli.

Polymeric nanoobjects

Templating of polymeric nanoobjects has become another research focus in the group. Particular interest lays in the preparation of shape anisotropic nanoobjects, branched nanoobjects, and block nanorods. Applications ranging from photovoltaics to nanoswimmers are explored.

Polymers for energy storage

Rechargeable lithium-metal batteries (LIBs) are one of the most promising technologies for future electrochemical energy storage devices. In recent years, we have become interested in polymers contributing to LIBs. These include:

- sulfur-containing polymers as cathode materials for lithium-sulfur batteries,
- polymer electrolytes for solid-state batteries,
- and radical containing polymers for organic radical batteries (ORBs).

Selected papers

- 🗈 W. J. Chung, J. J. Griebel, E. T. Kim, H. Yoon, A. G. Simmonds, H. J. Ji, R. S. Glass, J. J. Wie, N. A. Nguyen, B. W. Guralnick, P. Théato, M. E. Mackay, Y-. E. Sung, K. Char, J. Pyun; The use of elemental sulfur as an alternative feedstock for polymeric materials; Nat. Chem. 5 (2013) 518.
- 2 F. D. Jochum, P. Théato; Temperature- and light-responsive smart polymer materials; Chem. Soc. Rev. 42 (2013) 7468.
- 3 K. A. Günay, P. Théato, H-. A. Klok; Standing on the shoulders of Hermann Staudinger: Post-polymerization modification from past to present; J. Polym. Sci., Part A-1: Polym. Chem. 51 (2013) 1.
- A. Das, P. Théato; Activated Ester Containing Polymers: Opportunities and Challenges for the Design of Functional Polymers; Chem. Rev. 116 (2016) 1434.
- 5 J. Shang, X. Le, J. Zhang, T. Chen, P. Théato; Trends in polymeric shape memory hydrogels and hydrogel actuators; Polym. Chem. 10 (2019) 1036.

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Composition of the team

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Patrick Théato is a full professor in polymer chemistry at the Institute for Chemical Technology and Polymer Chemistry at the KIT since 2017. He studied chemistry at the University of Mainz, Germany, and the University of Massachusetts, Amherst, USA, and obtained his doctorate degree from the University of Mainz in 2001. After postdoctoral studies at Seoul National University supported by Feodor Lynen Postdoctoral Research Fellowship and at Stanford University, he completed his Habilitation at the University of Mainz in 2007. From 2009 to 2012 he held a joint appointment at Seoul National University, Korea, within the WCU program. In 2011, he accepted a prize senior lectureship at the University of Sheffield, UK. From 2011 to 2017 he was an associate professor (W2) for polymer chemistry at the University of Hamburg, Germany. Since 2014 he has held an adjunct professorship at Seoul National University, Korea. Since 2017 he is full professor (W3) at Karlsruhe Institute of Technology (KIT), Germany.

His team works in close collaboration with industry and partner universities in America, Europe, and Asia.

Polymeric materials

Polymeric Materials and Complex Characterisation Techniques Prof. Dr. Manfred Wilhelm

The group focuses on polymeric materials, their molecular structure, and the resulting macroscopic material properties. In addition, the enhancement of characterisation techniques builds the base of our research. The research activities can be divided into four main topics: (a) synthesis of model polymers including characterisation, (b) development in rheology, (c) the development of simultaneous characterisation methods and (d) use of polymeric materials in new application fields.

The correlation of the molecular structure of a polymer with its macroscopic application properties builds the scientific basis of the research in the group of Prof. Wilhelm. One important part is the synthetic capabilities of the group allowing to create well-defined polymers with variable composition of monomers to achieve a defined morphology and controlled topology, e.g. long chain branching. This offers the possibility to study the influence of the molecular structure on resulting materials and applications properties (Fig. 1), e.g. the influence of the degree of branching or the optimisation of hydrogels for the use in desalination or construction of an osmotic motor. On the other side, defined model polymers are a prerequisite for the enhancement and development of characterisation methods as they are needed in the calibration and testing procedures. In this way, the different research strands are highly connected to each other, and each strand benefits from improvements in the other areas

> Examples of home build flow cells to combine SEC/LAC and compact ¹H NMR spectroscopy



Synthesis and characterisation of special polymers

The synthesis of special materials and model polymers is the base for most research projects in our group. Most frequently anionic polymerisation, radical polymerisation, and emulsion polymerisation are used. The common aspects for all investigation are polymer-based materials and the influence of specific molecular properties on the resulting macroscopic, often mechanical material properties [1].

An important aspect of this strand are self-organizing polymer structures based on block copolymers. One actual example here is the influence of the topology and/ or morphology on the foaming properties of polymers and copolymers. A further part is the fatigue behaviour of polymers and the influence of tri- and diblock copolymers [4].

Another aspect of this research strand is the correlation of topology with the final melt properties. Here the effects of molecular weight, topology, and monomer type on the resulting properties have been studied, an actual project is the synthesis of comb polymers where either backbone or side chains consist of deuterated material offering the possibility to see the effect of the mobility of one selectable part in ¹H-NMR relaxation spectroscopy.

Fig. 2 Interaction of research strand synthesis with research strand rheology. Model polymers with defined molecular weight and topology are synthesized via anionic polymerisation. With these fully customizable model systems, the correlation between the molecular structure and the mechanical properties of the final material can be examined.

Molecular Structure Topology, Composition Structure









Fig. 1 General concept of our research: Molecular structure, more precise the monomer, the molecular weight, the distribution and the topology, and its correlation to the final material or even directly to applications.

A further important topic is the study of rubber materials where the influence of the filler material and the interface and the interaction between the matrix and the filler plays an important role. In addition, also composite materials (polymer + CNT or inorganic fillers) and emulsion are subjects of ongoing research.

Method development in rheology

Mechanical and rheological properties are commonly used to set processing conditions or design polymer applications. These material properties originate in the molecular structure of the polymer. Neither this structure, nor its connection to the resulting material properties is usually known in detail. Therefore, the correlation between mechanical properties and the molecular structure is a major issue in our research (Fig. 2). Fourier Transformation Rheology (FT-Rheology), extends the standard method of oscillatory rheology into the non-linear regime and was developed in our group. Recently, we introduced a new non-linearity parameter Q. Examples of its use can be found in literature.

The parameter Q is extremely sensitive to the topology of polymers, one can distinguish e.g. linear and star polymers. It also reflects strongly the influence of longchain branching even at extremely low concentrations of branching points [1]. This is an important ability because the influence of long-chain branches on the processing properties is dramatic, but their detection and quantification is complicated via alternative methods. In addition, other material classes benefit from our developments in non-linear rheology, too. Examples are filled rubbers and emulsions.



Development of coupled methods

Rheology addresses macroscopic mechanical properties of polymers. However, additional information on the molecular scale is often necessary for a full interpretation of rheological results. Therefore, the simultaneous use of rheology with other methods is highly beneficial. So far, we have coupled small-angle X-ray scattering (SAXS), dielectric spectroscopy, and low-field ¹H-NMR relaxation spectroscopy with rheology [3]. As a new project, we currently are developing the combination of rheology and IR spectroscopy.

Another focus in this topic is the development of coupled methods based on size exclusion chromatography (SEC) to achieve a simultaneous characterisation of size and chemical information. The main idea is an excellent illustration of the strong interaction of our different research strands: As we intend to synthesise precisely defined model polymers, we also need methods allowing us to characterise all molecular parameters of a polymer (Fig. 3). The first axis, the average size and the size distribution can be examined with SEC, the chemical identification and average monomer composition can be clarified either with IR-Spectroscopy or NMR-Spectroscopy. The last dimension, special functional groups or end groups, is measured via the assistance of IR-QCL-Lasers (quantum cascade laser). This offers the possibility to identify and quantify also rare groups, e.g. end groups or branching points. Only the combination of these three methods in one combined technique offers the possibility of correlated information, e.g. the change in composition or branching degree as a function of molecular weight. FT-IR-SEC is developed to a final state, the combination with an IR-Laser (QCL) is already working, and SEC-NMR is developed with the use of a 80 MHz permanent magnet [2], but both latter methods are still a matter of further enhancements. Currently, we can detect one carbonyl-group in a polymer of 500.000 g/mol, and further improvements in sensitivity, especially for accessible functional groups, is still under development.



▲ Fig. 3 3-dimensional spectroscopy is needed for full characterisation of polymers. Molecular weight distribution, chemical composition and functionalisation (endgroups) are spanning up a 3-dimensional parameter space containing the full information about a polymer molecule.

Polymeric materials in new applications

In this strand, we realise new application ideas for polymers. Prominent examples are new applications for hydrogels normally used in diapers or for water delivery in agriculture. We showed that they can be used for water desalination or for the construction on an osmotic motor [5]. Both applications are based on the capability of hydrogels to change the ion concentration in electrolytes.

Another example is the entropy wheel, where polymeric elastomers are used to build a machine that transforms waste heat into mechanical energy.

The basic concept behind this is the usage of the change in the restoring force of elastomers under strain by ongoing temperature changes. A partially heated wheel starts to rotate, so it transfers heat to mechanical energy, and works for very low heat differences of only 50° C. This allows heat recovery also for cooling liquids at low temperature, which is not possible with currently available methods.

Selected papers

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- Mechanical Properties during Hydrogel Synthesis; Adv. Sci. 9 (2022) 2104231.
- fatigue behavior of solid polystyrene/polyisoprene di-and triblock copolymers; Macromolecules 53 (2020) 5572.
- Monitored by Time-Domain NMR; Macromol. Mater Eng. 305 (2020) 2000174.



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🗈 M. Abbasi, L. Faust, M. Wilhelm; Molecular origin of the foam structure in model linear and comb polystyrenes: II.

C. Fengler, J. Keller, K.-F. Ratzsch, M. Wilhelm; In situ RheoNMR Correlation of Polymer Segmental Mobility with

 V. Hirschberg, L. Faust, D. Rodrigue, M. Wilhelm; Effect of topology and molecular properties on the rheology and 5 A. Jangizehi, C. Fengler, L. Arens, M. Wilhelm; Synthesis of Superabsorbent Poly (vinylamine) Core-Shell Particles

Composition of the team

Head: Prof. Manfred Wilhelm Group leaders: Dr. Nico Dingenouts Dr. Christopher Klein

Manfred Wilhelm is a full professor in "Polymeric Materials" at the Institute for Chemical Technology and Polymer Chemistry at the KIT since 2006. He studied Chemistry in Mainz and Toronto. Afterwards, he did his Ph.D. in solid state NMR with Prof. H.W. Spiess (MPI, Mainz). During the Ph.D. he spent one year at the University of California, Santa Barbara, USA. After the Ph.D. he did a postdoc at the Weizmann Institute in Israel, then returned to the Max Planck Institute for Polymers in Mainz. After his Habilitation in 2001 he became associate Professor in 2004 at TU-Darmstadt.

Manfred Wilhelm received a number of prizes including the Reimund Stadler Prize (2001) and the nomination of the Neo Award in 2012. His scientific ideas have reached the general public through different media: TV ("Sendung mit der Maus, Wissen vor Acht", ARD) and newspapers, e.g. Frankfurter Allgemeine Zeitung and Neue Zürcher Zeitung. He was offered a chair at the Bayreuth University (2016) as well as a directorship at the IPF in Dresden (2017). He served the faculty as vice dean and dean from 2015-2021 and became full member of the Academy of Sciences and Literature in Mainz 2021.

Insight

Polymeric Materials and Complex Characterisation Techniques Prof. Dr. Manfred Wilhelm



▲ Assembly of the different parts and pieces that are needed for the Rheo-NMR combination.

Unique Rheo-NMR set up as developed in our group over 20 years. Combining advanced Rheology and time domain ¹H NMR relaxometry.

▼ Experimental set up to combine size exclusion chromatography (SEC) and bench top ¹H NMR spectroscopy.









▲ Time domain ¹H NMR allowing to quantify molecular mobility within the 1–3 nm range.

▼ Sample test tube as inserted into the time domain 1H NMR spectrometer.

Photos: M. Breig, KIT

Spectroscopy of surfaces and interfaces in energy conversion devices

Applied Spectroscopy Prof. Dr. Clemens Heske

The research approach of our team relies on spectroscopic methods to study surfaces and interfaces in energy conversion devices. We utilize electron and x-ray spectroscopy, both in the lab as well as at third-generation synchrotron light sources around the world. With this "tool chest", we team up with national and international partners to unravel the secrets of surfaces and interfaces in a large number of different materials classes and devices.



Energy conversion devices

The function, quality, and, ultimately, efficiency of devices for energy conversion crucially depend on the electronic and chemical properties at their surfaces and interfaces. Often, such devices are initially optimized by a trial-anderror approach.

However, after this initial optimization, an understanding of the processes at such interfaces and surfaces becomes increasingly important, and hence further optimization is greatly facilitated by in-depth characterization.

Cutting-edge characterization

The central focus of our group is to utilize and develop sophisticated characterization methods, and to team up with other research groups and institutes as well as industrial partners who focus on the production and optimization of energy conversion devices.

In these collaborations, the team uses a combination of lab- and synchrotron-based spectroscopies to characterize the involved interfaces and surfaces. This scientific approach, which significantly extends beyond simple (standardized) characterization techniques, delivers much deeper insights into the investigated material system. This, in turn, helps to optimize the materials and devices under study.



Fig. 1 Picture of the optical hutch at the X-SPEC beamline [A], showing the double crystal monochromator (2000 – 15000 eV) on the right and the VLS plane grating monochromator (70 – 2100 eV) on the left.

◀ Bird-eye view of the experimental endstations at the X-SPEC Beamline.

International network

The group at KIT is embedded in a larger research network including the Institute for Photon Science and Synchrotron Radiation (IPS) at KIT Campus North, the University of Nevada Las Vegas (UNLV), and the Advanced Light Source (ALS) at the Lawrence Berkeley National Lab. This network strengthens the experimental capabilities of the group by having access to instrumentation available at the different locations and involves the group in research activities on a large variety of different applied systems, ranging from thin-film solar cells [1, 2], over devices for electrochemical hydrogen production, fuel cells, and hydrogen storage, to batteries and catalysts (e.g., [2])).

Liquids and method development

Furthermore, the team also pursues more fundamental questions related to electronic structure (e.g., of liquids), as well as instrument and method development. These efforts mainly focus on the field of soft X-ray spectroscopy. Our group investigates liquids, solutions, and gases, which requires sophisticated experimental setups that separate these samples from the ultra-high vacuum (UHV) environment of the measurement chamber and beamline with an ultra-thin membrane.

The X-SPEC beamline, in situ and operando spectroscopy, and the RIXS map

Over the last decade, the network has gained extensive experience developing such experimental setups and has built the X-SPEC beamline [4] at the KIT synchrotron (Fig. 1). The X-SPEC beamline has a world-wide unique energy range from 70 to 15,000 eV, combining soft and hard x-ray spectroscopy techniques. It offers experimental stations specialised for UHV as well as *in situ* and *operando* environments. The operando experimental station is a further development of the SALSA endstation (Solid And Liquid Spectroscopic Analysis [5]), which our network developed for operation at the ALS. A crucial ingredient is the design of high-resolution, high-transmission soft x-ray spectrometers with transmissions surpassing other state-of-the-art spectrometers by more than an order of magnitude. This makes it possible to collect data in a "RIXS map" [RIXS: Resonant Inelastic (soft) X-ray Scattering], as shown for a perovskite solar cell sample in Fig. 2 $[2^{h}]$.

In addition to the generation of valuable insight into the fundamental properties of material systems, the developed experimental setups and measurement approaches form the foundation for the in situ investigation of applied systems, such as the ones described above. These often require the study of liquids, gases, or surfaces in an ambient environment, e.g., at a solid/liquid or solid/gas interface. In Fig. 3 (right), an example of an experimental setup for the investigation of a solid/liquid interface is shown, together with RIXS maps recorded from liquid H₂O and D₂O (left).



▲ Fig. 2 Schematic structure of a perovskite solar cell sample investigated by RIXS (left) and a nitrogen RIXS map of such a sample (right). Excitation and emission photon energies are shown on the ordinate and abscissa, respectively [2].



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- 🗈 L. Weinhardt, M. Blum, O. Fuchs, A. Benkert, F. Meyer, M. Bär, J.D. Denlinger, W. Yang, F. Reinert, C. Heske; RIXS investigations of liquids, solutions, and liquid/solid interfaces; J. Electron Spectrosc. Relat. Phenom. 188 (2013) 111.



▲ Fig. 3 Left: RIXS maps of liquid H₂O and D₂O. Right: Experimental setup to investigate a solid/liquid interface using soft x-ray spectroscopy.



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Composition of the team Head: Prof. Clemens Heske Group leaders: Dr. Lothar Weinhardt Dr. Dirk Hauschild

In 1998, Clemens Heske received his "Dr. rer. nat." (Ph.D.) in Physics from the University of Würzburg, Germany. After two years as a postdoctoral fellow at the Advanced Light Source, Lawrence Berkeley National Laboratory, he became a "wissenschaftlicher Assistent" at the University of Würzburg and completed his German Habilitation in Experimental Physics in December of 2003. In 2004, he joined the University of Nevada Las Vegas (UNLV) Chemistry Department as an Associate Professor for Materials/Physical Chemistry, and was tenured in the summer of 2007 and promoted to Professor in the summer of 2009. In the fall of 2011, Clemens Heske was additionally appointed one of the directors of the ANKA Synchrotron Radiation Facility and the Institute for Photon Science and Synchrotron Radiation (IPS) at the Karlsruhe Institute of Technology (KIT), Germany, and as a Professor in KIT's Institute for Chemical Technology and Polymer Chemistry (ITCP).



Applied Spectroscopy Prof. Dr. Clemens Heske





- ▲ View of the sample environment inside the MFE lab's surface characterisation system.
- ◀ Surface characterisation system in the Materials For Energy (MFE) lab.
- ▼ Heatable gas-phase reactor for *in situ* studies.





Theoretical heterogeneous catalysis

Applied Computational Catalysis Prof. Dr. Felix Studt

The Studt group is working with the theoretical description of catalytic processes at the atomic-scale. This involves guantum chemical calculations of reaction mechanisms as well as corresponding kinetic modeling.

The drive for sustainable energy solutions requires a strong focus on catalysis, as it is essential to be able to efficiently transform energy from one chemical form into another. The research of AK Studt aims at an understanding of how catalysts work at the atomic-scale. This understanding can be obtained through quantum chemical calculations. In the field of heterogeneous catalysis, these are usually based on density functional theory (DFT). DFT calculations can be used to obtain parameters like adsorption energies, chemisorption energies of intermediates on catalytic surfaces and reaction barriers. If these calculations are coupled with (micro-)kinetic models of surface processes an in-depth understanding of the factors that limit the catalysts performance can be achieved. Identifying key descriptors that ultimately determine the activity and selectivity of materials and establishing those descriptors for a variety of catalysts allows for the construction of volcano-shaped curves where the optimal catalyst is characterized by an intermediate bond strength of the reaction intermediates. This strategy allows for the fast computational screening of new catalytic materials and has been successfully demonstrated for (transition-)metal based catalysts where the first examples of computationally discovered materials emerged.

Energy-related catalysis

The main focus of AK Studt is the conversion of CO₂ and hydrogen to chemicals and fuels. The two main processes related to this are the conversion of CO₂ to methanol using transition-metal catalysts and the conversion of methanol to olefins, hydrocarbons and aromatics using acidic zeotypes. This route would allow to base the chemical industry on a renewable resource if hydrogen is obtained from electrocatalytic water splitting.

Electro-catalysis

In principle, CO₂ can also be converted in a direct electrochemical process to e.g. olefins. AK Studt is therefore also interested in the modeling of the electrochemical steps involved in this reduction. While this is inherently more difficult than modeling of thermochemical reaction steps, the computational hydrogen electrode model represents an elegant way of circumventing this.

Single Atom Catalysis

Another avenue explored by AK Studt is the employment of single atom catalysts which have represent the highest dispersion and have therefore the potential of being the most atom efficient heterogeneous catalysts achievable. This, however, can only be accomplished if an in-depth understanding of the activity and stability of these catalysts is reached. The correlation of these type of catalysts with their homogeneous counterparts is highly intriguing and allows to develop unifying concepts between these two disciplines.

Selected papers

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- 3 J. Amsler, B. B. Sarma, G. Agostini, G. Prieto, P. N. Plessow, F. Studt; Prospects of Heterogeneous Hydroformylation with
 - Supported Single Atom Catalysts; J. Am. Chem. Soc. 142 (2020) 5087. E. Z.-J. Zhao, S. Liu, S. Zha, D. Cheng, F. Studt, G. Henkelman, J. Gong; Theory-guided design of catalytic materials using
- scaling relationships and reactivity descriptors; Nat. Rev. Mater. 4 (2019) 792.
- Using Ab Initio-Based Kinetics; J. Am. Chem. Soc. 141 (2019) 5908.



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formation, fate and consequence for catalytic activity of Pt single sites on CeO₂; Nat. Catal. 4 (2020) 824.

E P. N. Plessow, A. Smith, S. Tischer, F. Studt; Identification of the Reaction Sequence of the MTO Initiation Mechanism

Composition of the team

Head: Prof. Felix Studt Group leaders: Dr. Phillip Plessow Dr. Dmytro Sharapa Dr. Jelena Jelic

Felix Studt studied chemistry at the Christian Albrechts University (CAU) in Kiel. He completed his PhD at CAU at the department of Inorganic Chemistry under the guidance of Felix Tuczek in 2005. He continued as a postdoc at the Center for Atomic-scale Materials Design at the Technical University of Denmark. Afterwards he moved to the SUNCAT Center for Interface Science and Catalysis (SLAC National Accelerator Laboratory/Stanford University) where he worked as a staff scientist. In 2016, he was appointed Professor of Applied Computational Catalysis at the Karlsruhe Institute of Technology.

Felix Studt is the author of more than 150 publications and has co-authored one text book in the area of heterogeneous catalysis.

Optical diagnostics of reactive and non-reactive flows

Optical Diagnostics Group Prof. Dr. Rainer Suntz

The research of the Optical Diagnostics Group is dedicated to the development and application of laser-diagnostic and tomographic methods to investigate reactive and non-reactive flows.



Soot formation

The formation and oxidation of soot is investigated by laser-based techniques to obtain information about soot particle concentrations, number densities, mean primary particle sizes, size distributions, and reactivities against oxidation in various configurations such as laminar and turbulent flames, combustion chambers, and the exhaust gas of combustion engines.



Turbulent flows

One-dimensional single-shot spontaneous Raman-scattering measurements are carried out in turbulent coflow diffusion flames. Instantaneous values of the mole fraction of all the majority species (CH₄(fuel), N₂, O₂, CO, CO₂, H₂O, H₂) can be detected simultaneously along the horizontal propagation direction of the laser beam. Two-dimensional maps of the averaged mole-fractions

Mixing of fluids in a jet-in-crossflow arrangement

The mixing behavior of fluids, e.g. in a Jet-in-Crossflow arrangement, is a further research topic. For this purpose, a fluorescent tracer is added to the jet of the wind tunnel whereas particles are added to both, the jet and the crossflow. Planar-Laser-Induced Fluorescence (PLIF) combined with Particle Image Velocimetry (PIV) are used to measure simultaneously two-dimensional concentration-



▲ Fig. 3 Normalized Variances < s's'> of the tracer-concentration s (left) and Reynolds-fluxes in axial (middle) and lateral (right) direction of the crossflow measured at 4.5 jet diameters above the base plate

 Fig. 2 Two-dimensional maps of the averaged mole-fractions of the fuel CH₄ as an educt and CO_2 as a product of a turbulent co-flow diffusion flame (core-flow: mixture of 20 % CH₄ and 80 % N₂; ring-flow: 100 % O2) obtained by spontaneous Raman-scattering [N. Ebersohl, Th. Klos, R. Suntz and H. Bockhorn, 27th Symposium (Inter-national) on Combustion, The Combustion Institute, Pittsburgh, 997 (1998)].

are given for the fuel CH_4 as an educt and CO_2 as a product species of the combustion process by successive measurements at different heights above the burner (Fig. 2). Using this technique, PDFs (Probability Density Functions) are directly obtained to describe the chemical reaction of turbulent reactive flows via an appropriate transport equation.

and velocity-fields. From these measurements 2d-maps of variances of the concentration of the tracer, Reynolds-fluxes, and Reynolds-stresses are obtained which reflect the enhanced flux of turbulent compared to laminar flows (Fig. 3). These quantities are of special concern for the development and validation of turbulence- and mixing-models.

of the crossflow [C. Cárdenas, R. Suntz, J. A. Denev and H. Bockhorn, Appl. Phys. B, 88, 4, 581 (2007)].

In situ planar laser induced fluorescence in catalysis

The conversion of NO with molecular hydrogen is observed in a single channel over a Pt/Al₂O₃ catalyst by in situ PLIF of NO with high spatial resolution (Fig. 4). This method opens new prospects for the development of appropriate models in catalysis.

Fig. 4 NO conversion X_{NO} with molecular hydrogen in a single channel over a Pt/Al₂O₃-catalyst by in-situ PLIF of NO [A. Zellner, R. Suntz und O. Deutschmann, Angewandte Chemie, 127, 9, 2691 (2015)].



Optical tomography

A fast Planar Optical Emission Tomograph (POET, Patent 10 2009 044 303, s. Fig. 1) has been developed to detect 2d-maps of up to two different chemiluminescent species (e.g. OH*, CH*) and the thermal radiation of soot in laminar and turbulent flames simultaneously. With POET we can obtain information about heat-release rates, equivalence ratios, and – in case of fuel rich combustion - positions of the soot formation with respect to the oxidation zone of flames. POET consists of 10 Kepler-telescopes surrounding the investigated object at different angles. The chemiluminescence signals

collected by the telescopes are imaged onto the face surface of an optical cable, respectively. The latter consists of 90 single fibres, which are arranged side by side in a single row. The opposite ends of these 10 cables/900 fibres are merged together in a light collector, which images the output signal of all the fibres used for the tomographic reconstruction onto a single image intensified CCD-camera. Single shot experiments are carried out in various specially shaped laminar premixed flat flames and laminar and turbulent diffusion flames (Fig. 5).



▲ Fig. 5 Simultaneously acquired, normalized tomographic reconstructions at 310 nm (OH*, left), 430 nm (CH* + soot, middle) and 750 nm (soot, right) measured at a turbulent ethylene/air diffusion flame 4 mm above the burner with an exposure time of 1 ms. The white dashed lines are the 25% isolines of distributions at 750 nm (left and middle) or 310 nm (right). [Th. Häber, H. Bockhorn, R. Suntz, energies, 13(9), 2335 (2020)].

Selected papers

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C. Cárdenas, J.A. Denev, R. Suntz, H. Bockhorn; Study of Parameters and Entrainment of a Jet in Crossflow Arrangement

🗈 A. Zellner, R. Suntz, O. Deutschmann; Two-Dimensional Spatial Resolution of Concentration Profiles in Catalytic Reactors

Composition of the team

Head: Prof. Rainer Suntz Group leaders: Dr. Thomas Häber Dr. Camilo Cárdenas Dr. Sui Wan

Rainer Suntz is an Adjunct Professor (Apl.) of Chemical Engineering at the Institute for Chemical Technology and Polymer Chemistry at the Karlsruhe Institute of Technology (KIT). He studied Physics at the University of Heidelberg and carried out his Diploma thesis at the Max Planck Institute for Nuclear Physics in Heidelberg. In 1989, he got a Ph.D. in Physical Chemistry in the group of Professor Dr. Jürgen Wolfrum. After spending two years in industry he went along with Professor Dr. Henning Bockhorn to Kaiserslautern and later on to Karlsruhe as the head of the division "Optical Diagnostics". In his group, he carried out his Habilitation in Chemical Engineering in 2000.

Rainer Suntz is appointed member of the committee "Particle Measuring Techniques" of the ProcessNet Initiative

Exhaust Gas Center Karlsruhe

Prof. Dr. Olaf Deutschmann Prof. Dr. Jan-Dierk Grunwaldt

The Exhaust Gas Center Karlsruhe offers a comprehensive service in the field of catalytic exhaust gas aftertreatment. Due to many years of successful research, the working groups of Prof. Dr. Olaf Deutschmann and Prof. Dr. Jan-Dierk Grunwaldt (both KIT) enjoy international recognition. The constant contact and cooperation with different research centers in Germany, Europe, and worldwide, as well as numerous well-known automobile manufacturers and suppliers, underline the relevance and reliability of our approach, equipment, and methods.

Founded in 2010, the Exhaust Gas Center Karlsruhe offers a variety of complementary expertise ranging from catalyst preparation and testing to in-depth characterization, modeling, and simulation of chemical reactions and transport phenomena (Fig. 1).

Fig. 1 Available expertise in the Exhaust Gas Center Karlsruhe.



Conventional and modern preparation and coating methods are employed to obtain different classes of catalysts for emission control: oxidation catalysts, three-way catalysts, NOx- and VOC- removal catalysts, as well as catalysts for soot oxidation and urea hydrolysis. Novel developed materials are evaluated in comparison to applied industrial catalysts on in-house built synthetic gas test benches for catalyst powders and coated honeycombs. Kinetic measurements are conducted with gas mixtures containing up to 20 different components, at gas hourly space velocities

Furthermore, the center owns or has access to various material characterization methods (Fig. 2): X-ray diffraction, X-ray photoelectron spectroscopy, UV-Vis, IR and Raman spectroscopy, IR thermography, thermal analysis, electron microscopy, etc. Detailed structural and mechanistic aspects in a time and spatially resolved manner are studied by advanced in situ / operando synchrotron-based techniques like X-ray absorption / emission spectroscopy and X-ray microscopy. Due to the penetration ability of



relevant for real applications at pressures up to 6 bar. Axially resolved profiles of species concentration and temperature can be obtained via a capillary / thermocouple inserted into selected channels of catalytic monoliths at the SpaciPro setup. For obtaining 2D concentration profiles in a catalytic flow cell, we operate the laboratory for CATalysis at High Temperature Laser ENvironment (CATHLEN), which consists of a non-invasive in-situ experiment utilizing laser induced fluorescence (LIF) as well as Raman spectroscopy.

hard X-rays, such operando experiments can be performed under reaction conditions similar to those used in real applications. In this way, the dynamics of catalyst structure can be directly correlated with its catalytic activity, and this information is exploited for rational catalyst preparation.

Fig. 2 In-depth catalyst testing and characterization.



▲ Fig. 3 Modeling and simulation.

Finally, our research activities focus on the development of multi-scale models for the description of exhaust catalysts from the atomic level, over meanfield averaged reaction kinetics, to reactor-scale models including the physical transport processes, e.g. complex flow and temperature distribution in monolithic structures (Fig. 3).

In addition to all available facilities, the efforts of over 40 staff members, including scientists, engineers and technicians, enable broad fundamental research, service activities, and an effective transfer of knowledge-based catalyst development and implementation.



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