Inauguration Workshop of the CAT-ACT Beamline for Catalysis and Radionuclide Research at KIT

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Figure 1: Participants of the CAT-ACT beamline inauguration workshop grouped in front of the KARA accelerator building.

On September 20 and 21, 2017, a two-day workshop dedicated to the inauguration of the new beamline for catalysis and radionuclide research (CAT-ACT) at the Karlsruhe Institute of Technology (KIT) synchrotron light source took place in Eggenstein-Leopoldshafen on KIT Campus North. The workshop was co-organized by the three KIT institutes which have conceived and are jointly operating the beamline—the Institutes for Chemical Technology and Polymer Chemistry (ITCP), Nuclear Waste Disposal (INE), and Catalysis Research and Technology (IKFT)—and highlighted research activities and access possibilities at two in-line experimental stations. More than 60 national and international delegates attended the two day symposium, which also featured oral contributions from industrial synchrotron users and, of course, researchers from Karlsruhe (Figure 1).

The workshop was opened by Johannes Blümer, head of KIT Division V "Physics and Mathematics," who set the scene and emphasized the importance of pursuing new and groundbreaking research at the beamline, even going beyond the dreams and expectations of the scientific communities profiting from the new opportunities at KIT. The director of the Institute for Beam Physics and Technology (IBPT), Anke-Susanne Müller, presented the two accelerator facilities, FLUTE (Far Infrared Linac and Test Experiment) and KARA (Karlsruhe Research Accelerator), operated by IBPT as synchrotron radiation sources and accelerator component test facilities, underlining the close connection between beam physics and applied research; e.g., in catalysis and actinide sciences.

The CAT-ACT project history and unique potential of the highly versatile beamline were presented by Jörg Rothe (INE) and Jan-Dierk Grunwaldt (ITCP). They acknowledged support from KIT, third-party funding, various Helmholtz programs, and many colleagues who made planning and installation of this collaborative research group (CRG) beamline possible—too many to be mentioned here in detail. The CAT-ACT wiggler source provides high photon intensity combined with an exceptionally broad energy range and high beam stability, and will offer experimental conditions complementary to those at upcoming high-brilliance light sources. The aim to synergize synchrotron-based technology with research activities in catalysis/chemical engineering and radionuclide/nuclear waste disposal research fields requires a highly specialized lab infrastructure that has been realized in close collaboration with the operating institutes in the immediate vicinity of the synchrotron—a special feature of KIT. Crucial requirements for such a multi-purpose beamline are the high degree of automatization and general "user-friendliness" enabling; e.g., quick switching between different energy ranges, experimental stations, and spectrometer setups. Hence, an encoder signal-based alignment protocol has been implemented in the initial commissioning phase by the beamline scientists who did—according to the statements of early beamline users—an excellent job. Scott Mowat of FMB Oxford—developer and manufacturer of the beamline optics—outlined in his technical presentation the challenges of the CAT-ACT optic design, pointing out that precision and stability of collimating and focusing mirrors, which used to be limiting factors in the earlier days of commercial beamline optic development, have tremendously improved over the recent years. In this sense, CAT-ACT is considered as a milestone in beamline manufacturing, offering state-of-the-art monochromator and mirror equipment allowing for:

- fixed beam positions at the sample stage at all possible optic configurations with and without mirrors;
- a broad photon energy range spanning from 3.4 keV to beyond 55 keV;
- variable focusing at the two end stations, ACT and CAT, at about 23 m and 28 m from the source, respectively.

Following completion of the CAT-ACT beam delivery system and licensing of the accompanying personnel safety and radiation protection infrastructure in summer 2015, first X-ray absorption spectra (XAS data) were successfully recorded by the end of the year 2015. Since then, the commissioning proceeded smoothly and thus a number of in-house results could already be presented at the workshop (e.g., by Tonya Vitova (KIT-INE), highlighting recent high-resolution (HR) XANES studies at the actinide M4,5 edges (Figure 2), Henning Lichtenberg and Anna Zimina (KIT-IKFT) presenting absorption spectra of catalysts recorded under demanding reaction conditions (e.g., 20 bar pressure) with the special infrastructure for catalytic studies at CAT) [1].

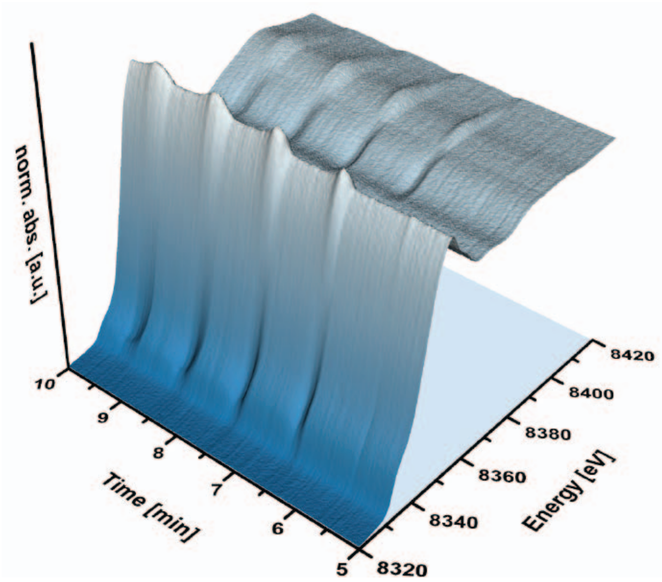

Figure 3: Structural changes in a Ni/Al_2O_3 catalyst during hydrogen drop-outs representing the fluctuating availability of resources during the "Energiewende"; each time hydrogen is removed from a H_2/CO_2 methanation gas mixture, the catalyst is slightly oxidized (cf. B. Mutz et al., Catalysts 7, 279 (2017). Published under Creative Commons Attribution 3.0 DE License).

Highlights of the workshop were presentations given by the invited guests. Ronald Frahm from Wuppertal University outlined, in his plenary talk "XAS and catalysis: An ideal symbiosis," the importance of synchrotron radiation for catalysis research since the early catalytic studies by van Nordstrand, emphasizing that synchrotron methods will be superior to laboratory XAS techniques for a long time. He also praised the progress of the activities in combining XAS and catalysis at KIT since the last "catalysis workshop" in 2010 in Karlsruhe (*SRN*, Vol. 24, p. 39 (2011)). Professor Frahm, a pioneer in the field of Quick-EXAFS (QEXAFS), also pointed out that the CAT-ACT optic design provides sufficient space for an additional QEXAFS monochromator, which would take time-resolved *operando* catalytic studies at KIT to a new level. The high potential of such studies was recently utilized as part of a collaboration between the Frahm group and KIT researchers to study the dynamic response of Ni-based catalysts to hydrogen drop-outs during methanation (cf.,

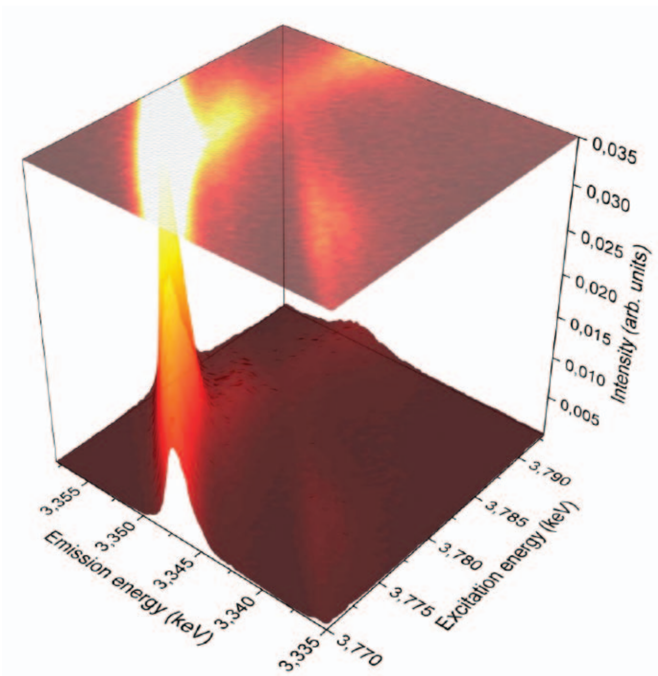

Figure 2: Pu M_5-edge 3d4f RIXS of PuO_2, recorded at the ACT station after commissioning of the high-resolution X-ray emission HRXES spectrometer.

Figure 4: The plenary speakers Ronald Frahm and Melissa A. Denecke, together with workshop organizer Jan-Dierk Grunwaldt and Nicolae Barsan inside the CAT experimental hutch during a beamline tour.

example depicted in Figure 3). The discussion following Frahm's presentation made clear that—beyond the challenge of providing suitable sample environments for experiments at synchrotron sources meeting relevant conditions of "real world" processes—time-resolution may be an increasingly important facet in future research activities.

Wolfgang Kleist (Ruhr University Bochum) demonstrated, during his talk, how X-ray absorption spectroscopy can be used as a tool to unravel the coordination environment of catalytically active metal centers in metal-organic frameworks, a relatively new and promising class of catalysts. The importance of XAS and related techniques in catalysis research was also highlighted by Christoffer Tyrsted from the internationally operating catalyst and engineering company Haldor Topsoe A/S. He showed that, since the pioneering work by Bjerne Clausen at Haldor Topsoe, XAS has evolved into a key technique in catalysis research, emphasizing that CAT-ACT also opens promising perspectives regarding detection setups combining XAS with complementary techniques like XRD, Raman, or IR spectroscopy. Synchrotron-based techniques allow researchers to follow the dynamic behavior of Cu-chabazite catalysts, an important system for selective catalytic reduction of NO_x by ammonia in exhaust gas after treatment, as shown by Martin Høj (Technical University of Denmark). The high potential of XAS with respect to energy-related catalysis was emphasized by two researchers from KIT: Silke Behrens (KIT-IKFT), who specializes in various synthesis and characterization techniques for the preparation of highly active catalysts, gave a talk focusing on catalysts for direct synthesis of dimethyl ether (DME) from synthesis gas, while Peter Pfeifer (KIT-IMVT) provided fascinating insights into the highly relevant topic of power-to-X technologies (i.e., transforming renewable energy into chemical energy) from a process engineering point of view. Nicolae Barsan (University of Tübingen), an expert in chemical sensing with extensive experience in *operando* characterization, added a colorful facet to the workshop program by reporting on *operando* XAS and DRIFTS (infrared) studies on semiconducting metal oxides for gas-sensing applications.

Melissa A. Denecke, scientific director at Manchester University's Dalton Nuclear Institute and former leader of synchrotron activities at KIT-INE, recapitulated, in her plenary talk entitled "X-ray spectroscopy and the nuclear fuel cycle," how XAFS spectroscopic/microscopic techniques increasingly have contributed, in the past decade, to various aspects related to nuclear fission energy, providing element-specific radionuclide speciation information, beginning with uranium mining and lastly covering spent fuel characterization, nuclear waste disposal, decommissioning, and remediation issues—the latter aspects highly relevant in the German context, where phase-out of nuclear electricity generation until 2022 was decided after the Fukushima incidence. Professor Denecke emphasized the urgent need to provide and further develop dedicated laboratory infrastructures enabling sample treatment and XAS data acquisition for radioactive materials, presently being realized only at a few synchrotron light sources worldwide due to difficult licensing and material safeguard procedures. Tobias Reich, head of the Institute for Nuclear Chemistry at Mainz University, covered the important aspect of how argillaceous geological formations, which are currently considered by several countries to host a future underground repository for highly radioactive nuclear wastes, may act as efficient sorbents for radionuclides. The group of Professor Reich was able to demonstrate that highly radiotoxic plutonium isotopes hypothetically released from a geological repository (following groundwater influx and canister and waste matrix corrosion) will be stabilized in-diffused into an Opalinus clay barrier as sparingly soluble Pu(IV) species through interaction with redox active mineral phases—a process which can be directly followed by spatially resolved Pu L_3-edge XANES and EXAFS spectroscopy. Although nuclear safety and waste disposal issues dominate the field of XAS applications at the existing dedicated end stations for radioactive materials, those radionuclides belonging to the actinide (5f) element series still pose a tremendous challenge for material scientists and theoreticians alike from a basic science point of view, as the contribution of their 5f-electrons in chemical bonding is often manifested in unusual physical properties or chemical behavior. That XAS is an ideal tool to illuminate such fundamental aspects was impressively demonstrated in the talk given by Nicola Magnani from the European Commission's Joint Research Centre at Karlsruhe, who reported on synchrotron radiation studies of the heavy-fermion superconductor $PuCoGa_5$. The scientific program was finalized by a contribution from Sumit Kumar from Bhaba Atomic Research Centre in Mumbay, India, currently holding an Alexander-von-Humboldt fellowship at KIT, who discussed actinide speciation studies in living marine organisms.

The opening workshop was wrapped up by a discussion about challenging future experiments at CAT-ACT, access routes to the experimental stations through collaborations or contractual cooperation, and future initiatives for possible upgrades. The workshop was preceded by a tutorial course on XAFS data analysis, where about 25 participants gained and intensified skills in XANES and EXAFS data treatment, creating opportunity for potential new users.

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Reference

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