Content

PREFACE 2
FOCUS 4

FACILITIES FOR EUROPE
Free-Electron Laser at ELBE 6
The Rossendorf Beamline (ROBL) at ESRF 8
TOPFLOW thermal hydraulic test facility 9

RESEARCH
Thermal fluid dynamics: 10
Two-phase flow - experiment and simulation

DYN3D - advanced reactor simulations in 3D 13

Magnetic flow control in metal casting and crystal growth 15

Cosmic magnetism in the lab 17

The reactor pressure vessel - a strong barrier to retain corium 19

How reactor pressure vessels grow old 20

Design of a photo-neutron source for time-of-flight measurements 22
at the Radiation Source ELBE

Bacteria and nuclear waste disposal 24

Self-organizing surface layer proteins as building blocks 26
for innovative nanomaterials

How mineral surfaces retard uranium migration 28

Environmental impact of depleted uranium 31

XAFS investigation of actinide species under controlled redox conditions 34

FACTS & FIGURES 36

Cover picture: The picture shows the result of a Computational Fluid Dynamic (ANSYS CFX®) calculation on the mixing of a de-borated slug of coolant (indicated in green) with the ambient coolant (indicated in red) in the reactor pressure vessel of a pressurized water reactor. The less dense slug stratifies in the upper part of the cold leg and flows around the core barrel in the top region of the downcomer. Mixing takes place on the way downwards to the core and mitigates the reactivity inserted.
Photo left: Rainer Weisflog
Preface

This volume of the Triennial Scientific Report highlights the scientific output of the FZD research program “Environment and Safety”, covering the years 2004 to 2006 and the first six months of 2007. It is one out of three volumes that are published this year for the first time. The first part of this report introduces the “Environment and Safety” program as well as the large-scale facilities that are used for research within this program. The second part consists of twelve articles on research projects that were conducted by scientists of the Institute of Safety Research and the Institute of Radiochemistry.

The last eighteen months were characterized by an intense discussion about the future of the Forschungszentrum Dresden-Rossendorf (FZD). In meetings and seminars we debated about our topical status and our future, asking questions like: “What are our future scientific objectives?” “Which research methods and facilities are required in order to reach those goals?” “How can new research activities be funded and who are our future cooperation partners?”

The Institute of Safety Research of the FZD is represented in many important European Networks and Integrated Research Projects, e.g. in the Sustainable Nuclear Energy Technology Platform which was launched in Brussels in September 2007. This platform aims at bringing together researchers and industry to define and implement a strategic research agenda and a corresponding deployment strategy. For the Environment and Safety program collaboration partners from scientific institutions and from European industry are of utmost importance. Just recently, the Institute of Safety Research and AREVA-NP Erlangen have signed an agreement on joint research projects. Workshops about the simulation of multi-phase flows have taken place in Dresden regularly, organized by the FZD together with ANSYS Germany GmbH. The TOPFLOW facility of the FZD is constructed especially for complex multi-phase flow experiments. The results of these experiments have been transferred into so called CFD (Computational Fluid Dynamics) codes like CFX® developed by ANSYS. Moreover, European industry makes use of the TOPFLOW facility for experiments regarding the safety of nuclear power plants.

Research on actinides, like uranium, plays the central role at the Institute of Radiochemistry of the Forschungszentrum Dresden-Rossendorf. As only few groups in Europe work on this special field of chemistry, close collaboration with the small scientific community is essential, in particular because radioecology addresses problems of immediate importance for society. Transport mechanisms of actinides affect questions which must be solved on the European level, e.g. the final disposal of waste from nuclear power plants. Naturally, the same is true for research on the safety of nuclear power plants.
The Rossendorf Beamline at the European Synchrotron Radiation Facility (ESRF) in Grenoble/France has proved to be an attractive experimental site for radiochemists from all over the world, because for many years it was the only synchrotron beamline where radioactive samples could be handled. The Rossendorf Beamline has guaranteed fruitful collaboration projects with many well-known European research institutions, partly financed by the European Union. For example, the Institute of Radiochemistry is actively engaged in the European ACTINET Network, where the training of young scientists is of special relevance. Furthermore, new cooperation projects with Japanese institutions like the University of Tokyo have started within the last few months.

Two prestigious prizes were awarded to FZD scientists of the Environment and Safety program recently: Prof. Gert Bernhard, Dr. Gerhard Geipel and Dr. Samer Amayri received the Kurt Schwabe Prize of the Saxon Academy of Sciences in 2005 for their discovery of a new uranium complex in mining water. Dr. Hans-Georg Willschütz was awarded the Karl Wirtz Prize 2007 of the Nuclear Technology Association (Kerntechnische Gesellschaft) for his research on the behavior of the reactor pressure vessel under severe accident conditions.

Finally, I would like to thank our partners in both the state and the federal government for their continued support, our national and international scientific cooperation partners for many successful joint research endeavors and, last but not least, the entire staff of the FZD for their dedicated work for this fine institution.

Prof. Dr. Roland Sauerbrey
This program is dedicated to the protection of humanity and the environment against industrial risks. For this purpose, our research aims at assessing and enhancing the safety of industrial plants and improving the efficiency and sustainability of the processes involved. Results from our research are applied to nuclear facilities of current and future designs as well as to installations in the process industries. It is the particular goal to assess and minimize the risks related to the nuclear fuel cycle. This includes the relics of former uranium mining, the power generation in nuclear reactors, and the final geological disposal of the radioactive waste. Many of the scientific methods and tools used in nuclear engineering can be successfully adapted to non-nuclear fields, for example to optimize industrial processes by customized flow control.

The Environment and Safety program is implemented by the Institute of Radiochemistry and the Institute of Safety Research. It includes the program areas Radioecology, Plant and Reactor Safety, and Thermal Fluid Dynamics. Whereas the Radioecology program area focuses on the identification of the chemical interaction and the mobility of radionuclides, especially of actinides, in the geo- and bio-spheres, our engagement in the Plant and Reactor Safety program area is directed towards the development of physical models used for accident analysis and the description of irradiation-induced ageing phenomena in reactor construction materials. Complementing this, the Thermal Fluid Dynamics program area studies transient multi-phase flows and magneto-hydrodynamic (MHD) phenomena to provide the basis for
advanced reactor simulation and optimization in process engineering, chemical industry, crystal growth, and metallurgy. The development and validation of tools for process simulation, accident analysis, and long-term safety assessment of the geological disposal of radioactive waste, with the emphasis on thermo- and fluid dynamic computation methods, constitute the framework for these program areas.

The research program is based on large-scale experimental user facilities operated by the FZD such as the Radiation Source ELBE, the Rossendorf X-ray beamline ROBL at the ESRF in Grenoble, and the Transient Two-Phase Flow Test Facility (TOPFLOW). Beyond that, close collaborations exist with the other research programs of the FZD, which are devoted to the Structure of Matter and to Life Sciences. These collaborations are for example related to the design of the pulsed photo-neutron source at ELBE and to the development of biological metal templates with modified magnetic properties for the Dresden High Magnetic Field Laboratory (HLD). In the fields of irradiation-induced materials ageing and surface layer characterization, we considerably benefit from the expertise at the Institute of Ion Beam Physics and Materials Research. Moreover, we contribute to the development of new macromolecules for nuclear medicine by our competence in laser spectroscopy.

The reactor safety and radioecology research is integrated in the German Alliances for Competence in Nuclear Technology (Kompetenzverbund Kerntechnik) and for Competence in Radiation Research (Kompetenzverbund Strahlenforschung), respectively. The magneto-hydrodynamic (MHD) project of the Institute of Safety Research forms a major part of the Collaborative Research Center (SFB) 609 funded by the German Research Foundation (DFG). In this SFB, the FZD closely collaborates with the Technische Universität Dresden and, for example, with the TU Bergakademie Freiberg on fundamental and applied MHD problems.
The migration behavior of radioactive heavy metal ions in groundwater aquifers is essentially determined by their interactions with mineral surfaces. The identification of the molecular species participating in the complex physico-chemical processes at the interfaces is a challenging task for modern actinide research.

Laser spectroscopic techniques have shown to be valuable tools for analyzing adsorbates on surfaces and for investigating the molecular processes occurring at the interfaces [1]. Particularly from the adoption of tunable infrared laser systems, a combination of surface selectivity and vibrational spectroscopic information can be expected [2]. Today, the most brilliant light sources for infrared light are accelerator-based light sources such as free-electron lasers (FEL). These lasers meet all the requirements for vibrational spectroscopic surface analyses due to their high spectral intensity, directionality, high degree of polarization, and coherence. The time structure of the pulsed light and the tunability over a broad wavelength range of interest complete the outstanding properties of this class of lasers.

At the FEL facility FELBE, an optical user laboratory is equipped for spectroscopic investigations of samples containing certain radioactive isotopes, obeying all aspects of radiation protection. As far as we know, this is unique for an FEL facility. The laboratory permits the application of advanced infrared spectroscopic techniques in actinide research [3]. A glove box provides the possibility to perform laser spectroscopic experiments on sensitive samples which have to be kept in an inert gas atmosphere excluding oxygen and carbon dioxide.

The experimental work focuses on vibrational spectroscopic investigations of actinide molecule complexes in solid samples using photothermal beam deflection spectroscopy. This technique combines the low detection limits of photothermal methods, vibrational spectroscopic information of the sample
under investigation [4], and surface selectivity [5]. Initial results of model samples containing low concentrations of actinide molecule complexes (UO$_2^{2+}$, NpO$_2^+$) were obtained by photothermal beam deflection [3].

Another experimental approach, which is substantially funded by the European Union and constitutes a joint research project with the Forschungszentrum Karlsruhe and the Paris National Institution of Higher Learning in Chemistry (ENSCP), uses the high intensity of infrared radiation to generate non-linear optical processes such as sum frequency generation to elucidate molecular structures occurring at well-defined interfaces in an aqueous medium [2]. The spectral information from the interfaces obtained by sum frequency generation is highly site-selective since neither in the bulk nor in nearly all crystal materials is a signal generated. Therefore, adopting a tunable FEL allows the acquisition of infrared spectra of radioisotope ions asorbed to a functional surface species. From these, spectra molecular structures of the surface complexes can be derived.

References
Andreas C. Scheinost

Synchrotrons provide extremely brilliant electromagnetic radiation across a wide range of energy, covering the infrared, visible, UV, and X-ray regions, which may be used for a large variety of spectroscopic and scattering techniques. Especially the hard X-rays with a small wavelength and high penetration depth provide versatile tools to probe physical and chemical states of matter in an almost unlimited variety of samples, from aqueous solutions and biological tissue to metal nanoclusters and thin films. The Rossendorf Beamline with its two experimental stations (Radiochemistry, Materials Research), built and operated by the FZD since 1998 is located at the most powerful synchrotron in Europe, the European Synchrotron Radiation Facility (ESRF).

The Rossendorf Beamline (ROBL) at ESRF

As a user facility, ROBL provides one third of its beamtime to the European research community via a proposal-based selection system managed by the ESRF. The rest of the beamtime is provided to FZD researchers and collaborating partners. The unique experimental facilities of ROBL are highly in demand by FZD and other research teams, including institutions like Max Planck, the ETH Zurich, and AMD (Advanced Micro Devices). From 2000 to 2002, ROBL was supported by the European Union within the framework of the “Access to Research Infrastructures” program. Since 2004, it has been part of the “ACTINET-EU Network of Excellence for Actinide Research”, providing its research facilities to all European member countries. ROBL beamtime is even sought after by research teams from overseas: currently a research contract is established between a consortium led by the University of Tokyo and the Japan Atomic Energy Agency (JAEA) to open ROBL for the Japanese Radiochemistry community.

To widen the experimental potential into the micrometer regime, we are currently planning a third station, where spectroscopy and diffraction will be implemented into a scanning X-ray microscope.

References
TOPFLOW thermal hydraulic test facility

Uwe Hampel, Helmar Carl

The multi-purpose thermal hydraulic test facility TOPFLOW (Transient Two Phase Flow Test Facility) is a device that is unique in the world for studying thermal hydraulic phenomena of steam-water two-phase flows at high pressures and temperatures. TOPFLOW has a maximum heating power of 4 MW and allows operation at pressures up to 7 MPa and temperatures of up to 286 °C in pipes and vessel geometries relevant to industry. One of its distinctive features is the advanced two-phase flow instrumentation, which includes high speed and IR cameras, wire-mesh sensors, phase-sensitive temperature sensors, and X-ray tomography. Furthermore, there is a large pressure chamber that serves as a pressurized laboratory for experiments in complex flow domain geometries. It allows operating thermal hydraulic test rigs of up to 7 m length and 2 m height in pressure equilibrium which enables optical and infrared flow observation through thin walls or glass windows.

TOPFLOW is devoted to basic and applied research on two-phase flow phenomena, which play a key role in many industrial fields such as chemical engineering (bubble columns, stirred reactors, electrochemistry), mineral oil processing, and nuclear technology. Currently, we are using TOPFLOW to qualify computational fluid dynamics codes (CFD) for two-phase flow simulations. Here, TOPFLOW is employed to derive appropriate closure laws for interfacial momentum and heat and mass transfer from experimental data.

TOPFLOW is the reference test facility of the German CFD Alliance on Nuclear Reactor Safety and is presently operated in the frame of two large projects. One comprehensive project is funded by the German Federal Ministry of Industry (BMWi). It deals with the provision of new experimental data for CFD code development and validation in reactor safety analysis. This includes two-phase steam-water flow in vertical pipes and horizontal flow channels, as well as in more complex geometries. The second project is concerned with the pressurized thermal shock phenomenon, which is a crucial reactor safety issue for aged reactor pressure vessels during emergency core cooling in a loss-of-coolant scenario. The project is a collaboration of seven industrial and scientific partners: Commissariat à l’Energie Atomique (CEA) France, Electricité de France (EDF), AREVA NP France, Institut de Radioprotection et de Sûreté Nucléaire (IRSN) France, Paul Scherrer Institute (PSI) Switzerland, ETH Zürich, Switzerland, and FZD. These two projects will completely occupy the experimental capacity of TOPFLOW until the year 2011.

References


Two-phase flows can be found in many industrial applications, as for example in nuclear installations and chemical process industries. They are characterized by a complex structure of the interface between the two phases. This structure of the interface results from the momentum transfer between the phases and determines heat and mass exchange. At the same time, the transfer through the interface is responsible for the evolution of the structure of the flow. Therefore, design, optimization and safety analyses require reliable tools for predicting the characteristics of the two-phase flows. The computer codes currently used, for example in the safety analysis of nuclear power reactors, are one-dimensional and based on empirical correlations valid only for special geometries, scales, and fully developed flows.

Computational Fluid Dynamic (CFD) codes have the potential to reflect the 3-dimensional nature of flow phenomena and consider the local flow properties. This approach could overcome the above-mentioned limitations. CFD codes are widely used for single-phase flows in the automotive or aviation industry, but have still to be qualified for two-phase flows. Due to the limited computational power it is presently not possible, and will not be within the next decades, to resolve the interfacial area of a two-phase flow completely for technically relevant flows. Instead, averaging procedures have to be used, which cause a loss of information regarding the interface. This information has to be supplemented by using so-called closure models. The main target of our work is to provide and validate such models and their interplay in simulations for relevant applications [1]. This task, however, requires detailed and reliable experimental data from multiphase flow experiments ranging from the study of separated flow phenomena in simple desktop flow channels to the testing of rigs with industrially relevant geometry, pressure and temperature scales, such as it is possible in the Rossendorf TOPFLOW facility.

Advanced measuring techniques applied in the TOPFLOW experiments

The Rossendorf TOPFLOW facility allows to conduct experiments at scales and operational parameters close to those found in the industry including nuclear power plants. The unique feature of TOPFLOW is that it is equipped with the most advanced measuring techniques that provide experimental data with a maximum of possible resolution in space and time. Among those techniques, there are several optical methods for measuring separated or diluted bubbly flows and wire-mesh sensors for dense bubbly flows, which are more relevant to practical applications.

The wire-mesh sensor technology was developed at the Institute of Safety Research and is successfully used here as well as in many other international research facilities. The wire-mesh principle is based on fast measurements of the electrical conductivity between the crossing points of two rectangular planes of wire electrodes. This allows to acquire cross-sectional phase fraction distributions at high speed (Fig. 1). A typical wire-mesh sensor used in TOPFLOW, capable of operating at pressures up to 7 MPa, provides flow structure details with a spatial resolution of 3 mm and at a rate of 2,500 frames per second. From the raw
data, characteristic multiphase flow parameters, such as time averaged local gas volume fraction distributions resolved with respect to the bubble size, can be obtained. Furthermore, velocity can be measured by using correlation techniques. Such detailed data are world-wide unique and used for CFD code development and validation by a number of national and international partners. The latest development is the preparation of a new imaging technique for flow measurement, namely ultra fast electron beam X-ray tomography. This new technique will not disturb the flow and provide flow data at temporal and spatial resolution equal or even better than wire-mesh sensors [2].

Qualification of Computational Fluid Dynamics (CFD) codes for two-phase flows
In adiabatic two-phase flows, the flow structure depends on the momentum transfer between the phases. Therefore, the momentum transfer between bubbles and the surrounding liquid is reflected in advanced CFD tools by closure models for the so-called bubble forces. From
investigations on single bubbles it is known that these forces strongly depend on the bubble size and one of them, namely the lift force, even changes its sign with increasing bubble size. This was confirmed for the first time for poly-dispersed flows by our experiments in vertical pipes. It was concluded from the experimental findings that the resulting separation of small and large bubbles has to be considered for a proper simulation of poly-dispersed bubbly flows. Based on these findings, the Inhomogeneous MUSIG model [3] was developed at the Institute of Safety Research and implemented in a numerical test solver which considers a number of different bubble size classes (Fig. 2). The model allows the consideration of a number of bubble classes for the mass balance as well as for the momentum balance. It was implemented into the CFX code jointly with ANSYS and is now part of the ANSYS CFX code. To prove the applicability of the Inhomogeneous MUSIG concept and the proposed closure models for more complex flows with pronounced 3D effects, experimental data was obtained for a flow around a half-moon shaped obstacle within a DN200 pipe.

Such advanced two-phase flow models also offer new insights into the character of industrially relevant flows. One example is related to the bubble columns, which are in wide use. It was shown that transitions from homogeneous to heterogeneous flow regimes are strongly connected to the lift force inversion [4].

Further experimental and theoretical studies around TOPFLOW are devoted to stratified flows, impinging jet configurations and two-phase flows with phase transfer. The experiments and the code development are harmonized within the German CFD initiative. TOPFLOW is the reference facility of this initiative. Most of the work is done in the framework of two projects funded by the German Federal Ministry of Economics and Technology (BMWi), but some special projects and tasks are also supported by other sources, as for example by the European NURESIM project. Currently, experiments on Pressurized Thermal Shock (PTS) phenomena are under preparation jointly financed by the French CEA, EDF, AREVA-NP, IRSN and the Swiss PSI.

References *

Project partners
1 ANSYS Germany GmbH, Otterfing, Germany
2 Gesellschaft für Anlagen- und Reaktorsicherheit, Garching, Germany
3 AREVA-NP, Erlangen, Germany/ Paris, France
4 Commissariat à l’Énergie atomique (CEA), France
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*In this report we quote mainly the most important papers that were published by FZD scientists and their partners.
DYN3D – advanced reactor simulations in 3D

Ulrich Rohde, Ulrich Grundmann, Sören Kliem

The computer code DYN3D

In nuclear reactors, transient processes with a significant reactivity insertion may occur, leading to an increase and 3D redistribution of the fission power density in the reactor core in a very short time. These transients can be induced by local changes in the temperature or density of the materials (fuel, moderator, absorber) or by the movement of the control rods. To ensure the inherent safety by a proper reactor design, and to estimate and minimize the consequences of hypothetical accidents, accident scenarios have to be modeled by adequate simulation tools. A best-estimate tool for simulating the dynamics of water-cooled reactors is the computer code DYN3D which was developed at the FZD Institute of Safety Research. It comprises a 3D-neutron kinetics model, a thermal-hydraulic calculation module, and a fuel rod model. The neutron kinetics model is based on the solution of the 3-dimensional two-group neutron diffusion equation by nodal expansion methods. By means of the thermal-hydraulic model, the density and temperature of the coolant are obtained under one- and two-phase flow conditions. By solving the corresponding heat conduction equations, fuel and cladding temperatures are calculated. These parameters are important for the thermal-hydraulic feedback on the neutron kinetics as well as for assessing the safety of the reactor. The nuclear cross sections in the neutron diffusion equations do not only depend on the feedback parameters, but also on the nuclide concentrations and the neutron spectrum, which change with the burn-up of the fuel. These dependencies must be taken into account providing corresponding cross section libraries. Fig. 1 shows the interaction of the neutron kinetics, thermal hydraulics, and neutron cross sections. The DYN3D core model was linked to thermo-hydraulic system codes [1] providing boundary conditions for the core, such as coolant inlet temperature and pressure and coolant mass flow rate distribution. System codes model the thermo-fluid dynamics of the primary and secondary circuit including all major components of the plant.

DYN3D is undergoing continuous development with respect to the improvement of physical models and numerical methods. Recently, a multi-group approach has been implemented in order to improve the description of spectral effects, which are increasingly important for mixed-oxide reactor core loadings of Light Water Reactors, but also for innovative reactor concepts. A neutron transport method was developed for DYN3D to overcome the limitations of the diffusion approximation. Within this approach, even a pin-wise calculation of the power distribution is offered. This was recognized internationally with strong interest [2, 3]. The extended DYN3D version was integrated into the European code platform NURESIM. An interface to the platform was developed, which is based on the NURESIM software environment SALOME. This allows linking DYN3D to other components of the platform, e.g., to advanced fluid dynamics simulation tools (CFD codes). In 2006, eight research projects related directly to the improvement, validation, and application of DYN3D were running or completed. Moreover, licenses for DYN3D, including commercial ones, were provided to 12 users in Germany and Europe. Within a PhD project, a version of DYN3D for dynamics studies of molten salt reactors (MSR), which belong to the 'Generation IV' concepts, was developed [4]. Analyses were performed for a number of specific MSR transients demonstrating the inherent safety of this reactor.

Analysis of a hypothetical boron dilution scenario

A reactivity initiated transient can be induced by the perturbation of the boron concentration in the core of a pressurized water reactor. Boron is added to the reactor coolant as a neutron absorber compensating the excess of reactivity in the fresh core at the beginning of the fuel cycle. Due to different mechanisms or

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Fig. 1: Scheme of interaction of the different models in the DYN3D code.
system failures, for instance as a consequence of a small break loss of coolant accident, slugs of low borated water can accumulate in the primary cooling system. During the start-up of the coolant circulation—after refilling the primary circuit with emergency cooling water—or by switching on the first main coolant pump (MCP), these slugs are transported into the reactor core causing a reactivity insertion by decreasing the amount of the neutron absorber. The boron concentration in the reactor core results from turbulent mixing along the flow path in the reactor vessel. In this case, mixing the de-borated condensate with borated water is the only mitigative mechanism to prevent re-criticality of the shut-down reactor and impermissible power excursion [5]. Computational Fluid Dynamics (CFD) methods are applied, allowing to ascertain the time-dependent boron concentration depending on space and time is crucial for the induced reactivity insertion. The boron distribution is obtained by modeling the mixing of the de-borated slug with the ambient coolant in the reactor by means of CFD. Fig. 2 shows the distribution of the boron concentration at the reactor core inlet when the boron content is at its minimum. DYN3D transient calculations were performed for different slug volumes. Considering the bounding scenario with a slug size of 36 m³, the reactor becomes promptly super-critical (maximum reactivity of about 2 $\$\$), leading to a very fast power excursion with a peak power of about 7000 MW (about twice of nominal power). Fig. 3 shows the time behavior of the reactor power. However, the power excursion is limited by the very effective Doppler feedback of the fuel temperature. The width of the power peak in time is only about 25 ms. Therefore, the integral energy release in the power peak is limited. The fuel temperature remains below 800°C, i.e., far below the melting point. There is local coolant boiling in the hottest fuel assemblies, but no cladding super-heating occurs. It can be concluded from the analyses that even in the case of a conservatively large volume of un-borated water, the safety criteria are met and the integrity of the core is not endangered. The investigations have shown large margins if a realistic approach to the modeling of the coolant mixing is applied.

Fig. 2: Boron concentration distribution over the core inlet cross section at the moment of minimum boron content.

**Fig. 3:** Reactor power (relative to nominal) during a boron dilution transient.

**References**


**Project partners**

1CEA (France)
2Vattenfall (Sweden/Germany)
3Fortum Nuclear Engineering (Finland)
4TÜV Süddeutschland (Germany)
5VGB PowerTech (Germany)
6AREVA (France/Germany)
7Nuclear Research Institute Řež (Czech Rep.)
Sven Eckert, Gunter Gerbeth

Electromagnetic fields represent an attractive tool to influence the flow and, in turn, the heat and mass transfer in electrically conducting fluids such as liquid metals or semiconductor melts. The most attractive feature of this magnetic control is that it works without contact, which is essential for hot and aggressive melts like liquid steel, aluminum, or silicon. In general, steady magnetic fields slow down the melt flow or suppress turbulent fluctuations, whereas alternating magnetic fields set the liquid in motion. Yet today, the challenge consists more and more in an inverse approach: a desirable flow field made possible by tailored magnetic fields. This inverse approach constitutes the key element of the Collaborative Research Centre (SFB) 609 at the Technische Universität Dresden, in which we are intensively involved.

**Experimental modeling and measuring techniques**

Experiments with hot metallic melts \((T > 600 \, ^\circ C)\) are difficult to carry out, expensive, and to some extent meaningless as long as the necessary measuring techniques for such melts are still lacking. Experimental data of the melt velocity are, however, indispensable for optimizing the melt flow. To fill in this gap, we developed a basic approach for performing cold liquid metal model experiments [1] together with velocity measuring techniques that allow measuring almost any interesting flow quantity up to about 300 °C. Our main model melt is the ternary alloy GaInSn, which is liquid at room temperature, but tin or lead based alloys with melting temperatures lower than 200 °C are also used. Today, ultrasonic Doppler velocimetry (UDV) has become the primary velocity measuring technique [2]. It provides an instantaneous velocity profile along the ultrasonic beam and can even operate through the wall of the melt container.

X-ray radiography presents a promising tool for visualizing processes in opaque metal melts in the future. For example, solidification processes or the motion of gas bubbles in liquid metals can be directly observed. As the Radiation Source ELBE at the FZD provides channeling radiation, a new X-ray lab was set up there. This will allow investigating a wide range of different alloys with both a high time resolution and an excellent signal-to-noise ratio. Fig. 1 shows solidification studies for a GaIn alloy performed with a microfocus X-ray tube. The brighter parts indicate the lighter gallium melt. A quantitative analysis of the brightness distribution allows reconstructing the thermosolutal convection in front of the solidified dendrites (Fig. 1b).

**Metal casting**

Today, new applications in the automotive and aviation industries are a challenge for the metal casting industry. In metal casting, the pouring process is known as a critical issue with respect to the casting quality. The high velocity at the beginning of the pouring process is a particular problem which is expected to have a significant influence on typical casting problems such as the entrapment of gas bubbles or inclusions into the casting units. This causes inhomogeneities in the microstructure which may significantly deteriorate the mechanical properties of the casting products.

Model experiments with the GaInSn melt revealed the critical importance of the incoming melt front. Fig. 2 shows snapshots of the initial moment of the pouring process in an industrial casting unit which is cut open here for the purpose of melt flow visualization. The melt front in the form of a breaking wave (Fig. 2a).
implies a high rate of gas bubble entrapment. This finally results in the inclusion of recognizable gas bubbles in the solidified microstructure in Fig. 3a. However, we were able to suppress the incoming breaking wave via magnetic control of the melt flow (Fig. 2b), which improved the microstructure (Fig. 3b). In a series of several hundred castings of aluminum alloys, the rate of casting failures was reduced by around four due to magnetic control of the pouring process [3].

Recent model experiments on gas-bubble liquid metal two-phase flows showed that applying a steady magnetic field does not always result in flow stabilization. Instead, it gave rise to velocity fluctuations in a limited range of magnetic field strengths [4]. In the future, this may qualitatively change the use of electromagnetic brakes in the two-phase nozzle region of the industrial continuous steel casting process.

Crystal growth from the melt
Today, single silicon or gallium-arsenide crystals are mainly grown from the melt. The quality, size, and growth rate of the crystals as well as the stability of the growth process strongly depend on the motion in the melt and related temperature fields. Typically, the melt is requested to show a flat solid-liquid interface, a low level of temperature fluctuations, and a controlled transport of oxygen or dopants. Magnetic fields are a powerful tool to provide this melt flow control. Scientists are particularly interested in gaining knowledge and control of the flow stability in order to keep the melt flow laminar, if possible [5]. As with larger melt sizes, the flow is always turbulent. However, tailored magnetic fields can be applied to reduce the temperature fluctuations significantly, for instance [6]. The flow topology, in turn, has a strong influence on the dopant distribution in the grown crystal [7].

References

Project partners
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Cosmic magnetism in the lab

Dynamo experiments, field reversals, and magnetic flow tomography

After the 1999 experiments in Riga and Karlsruhe had marked a breakthrough in dynamo science [1], many activities around the world aimed at studying magnetic field self-excitation in a variety of liquid sodium facilities. In Riga, a total of seven experimental campaigns have delivered a wealth of ever-refined measurement data on the kinematic and the saturated dynamo regime. Much effort was spent on the numerical simulation resulting in the recent quite accurate understanding [2] of this paradigmatic hydromagnetic dynamo (Fig. 1).

The codes developed for the simulation of the Riga dynamo were also applied to the French “von Karman sodium” (VKS) experiment. Our numerical findings [3] helped the VKS team to make their facility a working dynamo which even shows a sort of magnetic field reversals. We have analyzed this fascinating phenomenon within a simplified mean-field dynamo model which already exhibits many features of the earth’s magnetic field reversals in a realistic manner [4, 5].

As a spin-off of our dynamo activities we have developed the “Contactless Inductive Flow Tomography” (CIFT) [6] which has a promising potential for a fully contactless flow measurement in crystal growth and metallurgy. CIFT constitutes the main subject of the European project MAGFLOTOM, which was started in February 2007 and is coordinated by the FZD.

Experiments on the magnetorotational instability

The magnetorotational instability explains why stars and black holes can accrete matter from the gas disks surrounding them. Before being swallowed by the central object, the gas in such accretion disks has to lose its angular momentum. It has been shown that the molecular viscosity in the disks is much too small to explain the needed angular momentum transport. Turbulence could well do the job, but hydrodynamics teaches us that rotating flows with outward increasing angular momentum are stable. Here is the point where magnetic fields come into play as a catalyst to destabilize flows which are otherwise hydrodynamically stable.

The experiment PROMISE (Potsdam ROssendorf Magnetic InStability Experiment) is a joint project of FZD and...
Astrophysikalisches Institut Potsdam (AIP) and is supported by the Leibniz Gemeinschaft in the framework of its SAW program. PROMISE traces back to a proposal by Hollerbach (Department of Applied Mathematics, University of Leeds) and Rüdiger (AIP) to use helical instead of purely axial magnetic fields in order to make an MRI experiment feasible with reasonable effort. Fig. 2 shows the experimental set-up which basically consists of a Taylor-Couette cell of the liquid alloy GaInSn which can be exposed to a helical magnetic field. A typical feature of the MRI is visible in Fig. 3 and Fig. 4 which show that the instability in form of a traveling wave appears only in a finite interval of the applied magnetic field. More details about this experiment can be found in [7, 8]. Presently, we modify the set-up in order to make the transition between the stable and the unstable flow even sharper.

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1 Institute of Physics, Riga, Latvia
2 Delft Technical University, Delft, The Netherlands
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The reactor pressure vessel – a strong barrier to retain corium

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From the very beginning of the peaceful use of nuclear energy, a high safety level has been a major target within the development of nuclear power plants. The severe accident at Three Mile Island in 1979 provided a strong impetus for severe accident research during the last three decades. On the one hand, Three Mile Island showed that core meltdown scenarios are not hypothetical, but, on the other hand, it proved the safety margin of the pressurized water reactor system. The FZD focuses on the thermomechanical behavior of the most important safety barrier—the reactor pressure vessel (RPV)—during a postulated severe accident. In this case, the cooling of the reactor core cannot be re-established. The main question is: Can the reactor pressure vessel withstand a core meltdown if external cooling can be arranged? If the reactor pressure vessel cannot be excluded, potential failure modes and the time to failure have to be known in order to assess the possible loads on the containment. To solve these questions, a coupled thermal-mechanical finite element model was developed evaluating the transient temperatures in the melt pool and in the vessel wall as well as the viscoplastic deformation of the reactor pressure vessel [1 – 3]. The coupling not only allows considering the temperature dependence of the material parameters and the thermally induced stress, but it also takes into account the response of the temperature field itself upon the changing vessel geometry. New approaches are applied to the simulation of creep and damage. The mechanical damage for each wall element is incremented after each time step according to:

\[ \Delta D = \frac{\Delta \varepsilon^c_{ep}(T, t)}{\varepsilon^c_{ep}(0, t)} + \frac{\Delta \varepsilon^p_{ep}(T, t)}{\varepsilon^p_{ep}(0, t)} \cdot R_v \]  

In other words, the two possible strain increments—creep and plastic strain—are weighted against the according rupture strain. So initially the damage is zero; if a value of 1 is reached, the corresponding element is “killed”. The coefficient \( R_v \) considers the influence of the three-dimensional stress state according to a model proposed by Lemaitre [4], since high triaxiality makes the material brittle. The new models were widely validated by pre-test and post-test calculations on several internationally scaled in-vessel retention experiments, e. g., on the FOREVER tests, performed at the Royal Institute of Technology Stockholm [2, 3]. Figure 1 shows the calculated temperature field and damage distribution within the FOREVER vessel wall in comparison to metallographic post-test investigations.

The simulations of prototypic scenarios show that in-vessel retention of the melt could be possible by external flooding even at increased pressure and in reactors with high power. Two patents for passive devices were derived from the insights gained (Fig. 2): The first device (“Creep Stool Plates”) supports the lower head pole part of the reactor pressure vessel. It reduces the maximum mechanical load in the highly stressed area of the hot focus (see Fig. 1). In this way, it can prevent failure or at least extend the time to failure of the vessel, even in the case of a totally dry scenario. The second device triggers passive flooding of the reactor pit with the weight of the downward moving lower head, which relocates due to thermal expansion, creep, and plastic deformation (cf. Fig. 2).

References

Project partners
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How reactor pressure vessels grow old

The reactor pressure vessel (RPV) of a nuclear power plant (NPP) is the main barrier against the emission of fission products into the environment; it is not replaceable and it therefore limits the operation duration. The structural integrity of the reactor pressure vessel must be maintained throughout the operational time of a nuclear power plant under normal operation as well as under accident conditions. During operation, the mechanical properties of the pressure vessel are changed mainly due to irradiation with fast neutrons. This leads to an increase in hardness and an unwanted loss of ductility—in other words, the reactor pressure vessel grows old. The reasons for the ageing of RPV steels are complex and can be traced down to the nanometer scale: Irradiation with fast neutrons gives rise to displacement cascades in the crystal lattice accompanied by a substantial increase in the concentration of vacancies, which in turn serve as vehicles for long-term diffusional rearrangements of atoms. The resulting nanometer-sized defect-solute clusters are obstacles to the motion of dislocations, i.e., one-dimensional crystal defects acting as “quanta” of plastic deformation. In other words, the material hardens which is associated with a loss of ductility. The irradiation-induced clusters are thermodynamically metastable, they dissolve at temperatures of about 450°C and give rise to the recovery of the mechanical properties. In order to meet the highest standards of reactor safety and to develop advanced reactor concepts, a better and more detailed understanding of the property changes and the responsible microstructural mechanisms is required. The FZD contributes to these international efforts by experimental as well as computational means.

How to apply advanced fracture mechanics concepts to RPV integrity assessment

The Master Curve (MC) approach is a new fracture mechanical methodology developed to describe the fracture toughness behavior in the lower ductile-to-brittle transition (DBT) range of ferritic steels. This approach is rapidly moving from research to application, as it is used for assessing the integrity of the components and structures and thus of the reactor pressure vessel. The Institute of Safety Research contributes to the validation of the Master Curve concept and to its application to irradiated RPV steels [1]. The main question is whether the universal shape of the Master Curve is changed by neutron irradiation, which was investigated within a project funded by the German nuclear safety program [2]. The project was aimed at testing irradiated specimens of different RPV steels.

Fracture-toughness values $K_c$ at brittle failure of the specimens were measured for a reference RPV steel highly sensitive to neutron embrittlement. Fig. 1 shows the ductile-to-brittle transition temperature shift (up to 230 K) as a consequence of the neutron dose expressed in units of displacements per atom (dpa). Generally, the $K_c$ values follow the universal shape of the Master Curve for a fracture probability of 50% (lines in Fig. 1). We found that the physical background of the Master Curve approach is valid even for the RPV steel in the highly embrittled condition. The coincidence of the curves for the irradiated and annealed condition (IA) with the unirradiated reference (U) indicates the complete recovery of the toughness properties.

Taking snapshots of the size distribution of defect-solute clusters by SANS

Small-angle neutron scattering (SANS) is an advanced experimental technique which is unique in its capability to resolve the size-distribution of clusters at nanometer resolution while probing macroscopic volumes. Fig. 2 displays the measured size distributions for the investigated reference RPV steel.
We found that the volume fraction of defect-solute clusters increases with the dose, whereas the mean radius is approximately constant. For the highest dose, post-irradiation annealing treatments at different temperatures give rise to cluster dissolution. Despite the complexity of the dissolution process in steels, the dissolved fraction exhibits an Arrhenius behavior. It is assumed that the apparent activation energy of 1.2 eV is a fingerprint of the rate-controlling step of the dissolution process [3]. In order to confirm the relevance of these findings, the Vickers hardness was measured for the same samples which were used for the reported SANS measurements. Fig. 3 indicates a strong correlation between the increase in Vickers hardness induced by irradiation and the square root of the cluster fraction for both irradiated and annealed conditions. The re-irradiation behavior after annealing was investigated for a RPV weld material used for Russian VVER440-type reactors [4]. We have observed that a certain fraction of the irradiation-induced clusters coarsened and accumulated Cu during annealing. These Cu atoms are no longer available for the process of cluster formation during re-irradiation. As a consequence, the degree of embrittlement is less than for the first irradiation. This is important to know for evaluating the large-scale annealing treatments performed for a number of VVER440-type reactors in several countries and finally contributes to the highest standards of reactor safety.
Design of a photo-neutron source for time-of-flight measurements at the Radiation Source ELBE

At the Radiation Source ELBE, neutron time-of-flight experiments (nELBE) are envisaged for energy dispersive studies of neutron interactions with matter. Measuring neutron-induced reaction cross sections for fusion and fission reactors as well as for accelerator driven systems aiming at the transmutation of nuclear waste is one of the tasks to be pursued at the photo-neutron source. Additional applications arise in the field of astrophysics in improving the reaction rates of astrophysical nucleo-synthesis models. The short, ps-range pulses of the ELBE accelerator combined with a small neutron radiator volume allow the photo-neutron source to have an ultra compact design.

Pulsed neutrons are generated at nELBE when the ELBE electron beam hits the neutron radiator, which is made from liquid lead housed in a molybdenum channel inside a vacuum chamber. The electron pulse structure is transformed into neutrons with a similar structure. A flight path of about 4 m is sufficient to separate the neutron pulses from the bremsstrahlung flashes and secondary electrons and to determine the neutron energy by the travel time of their free flight.

The construction of the pulsed photo-neutron source was preceded by detailed simulations [1, 2]. The calculations proved that liquid lead is the most appropriate radiator material regarding neutronics as well as thermal criteria and that molybdenum is the best suited wall material in
the region where the electrons hit the radiator channel. The temperature distribution of in the radiator was simulated using computational fluid dynamics. Fig. 1 shows the temperature distribution that results from a 30 kW electron beam with 8 mm diameter hitting the Mo-radiator with a mean lead flow velocity of 1 m/s. Given the maximum electron current $I_e = 1$ mA of ELBE, the detailed Monte Carlo simulations predict a neutron source strength of $2.7 \times 10^{11}$ n/s and an average neutron flux of $1.5 \times 10^7$ cm$^{-2}$ s$^{-1}$ at the measuring position for an electron energy of $E_e = 40$ MeV. Using the new super conducting radio frequency (SRF) photo electron injector with a repetition rate of $f_e = 0.5$ MHz, the measurable neutron energy ranges between $E_n = 50$ keV and 10 MeV. We expect the energy resolution to be better than 1% for energies up to 5 MeV. When inserting a thermal neutron filter of polyethylene and cadmium in the collimator, the background resulting from the previous pulse can be reduced significantly (see Fig. 2) so that, for example, a signal-to-background ratio of $10^5$ is achieved for 2.5 MeV neutrons.

Compared to all existing high-resolution neutron beams nELBE has a very competitive luminosity in the accessible energy range. The advantage of such a small design is that the facility retains a high intensity with good resolution, whereas some other neutron sources lose a significant part of their source intensity advantage over nELBE when increasing their flight path in order to reach an energy resolution of better than 1%.

The design of the nELBE facility is a joint effort between the FZD Institutes of Safety Research and Radiation Physics and the Institute for Nuclear and Particle Physics of Technische Universität Dresden. The facility is now approaching completion and joined the efforts of the EFNUDAT (European Facilities for Nuclear Data Measurements) project. Ten institutions in seven European countries participate in this project, which is funded within the 6th framework programme (FP6) of the European Commission. The design of nELBE was supported by the German Research Foundation (DFG).

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All over the world, researchers are developing concepts for the disposal and final storage of nuclear waste in deep geological formations (Fig. 1). This is aimed at protecting the population and the environment from the risks caused by radioactive and hazardous nuclear waste. One European research facility to test the potential of crystalline bedrock for storing nuclear waste is the Äspö Hard Rock Laboratory (Äspö HRL), located on the southeastern coast of Sweden.

Microbes are widely distributed in nature and are capable of strongly influencing the migration of hazardous actinides in the environment. This has led to an increased interest in studies exploring the interaction processes between actinides and bacteria during the past several years. It is also reflected in the FZD research area “Radioecology” which, among other things, is dedicated to actinides in biological systems. Here, we have focused on exploring the manifold interaction processes between the actinide element plutonium and the sulfate reducing bacterium Desulfovibrio aespoeensis [1].

Our Swedish partners investigated the microbial diversity at Äspö HRL. The total number of microbes ranges from $1 \times 10^3$ to $5 \times 10^5$ cells ml$^{-1}$, whereas the number of sulfate reducing bacteria (SRB) was between $1 \times 10^1$ to $2 \times 10^4$ cells ml$^{-1}$. The ubiquitous SRB strain D. aespoeensis was isolated from the Äspö site at a depth of 600 m [2]. Microorganisms can interact with actinides directly (Fig. 1) as well as indirectly. For the first time, direct interaction processes between plutonium in different oxidation states (+6 and +4) with cells of D. aespoeensis were determined [1]. We found out that the reduction of Pu(VI) to Pu(V) by the bacteria leads to an increased dissolution of the cell-bound plutonium. In contrast to the release of Pu(V) from the cell surface into the surrounding solution, we observed an immobilization of Pu as Pu(IV) polymers by the biomass.

Plutonium is a hazardous actinide element which exhibits a complicated chemistry because it can exist in several oxidation states (+3, +4, +5 and +6) in an aqueous solution in environmental conditions. Therefore, studying the interaction of plutonium with microbes is challenging. Our work demonstrates that there is strong interaction between the two main plutonium species, Pu(VI) and Pu(IV), and the cells of D. aespoeensis. The distribution of the oxidation-states of plutonium is based on solvent-extraction experiments and was successfully confirmed by X-ray absorption near edge spectroscopy (XANES) performed at the Rossendorf Beamline (ROBL) at the ESRF and absorption spectroscopy measurements. Based on these results and on the work of Panak and Nitsche [3], we developed a model which describes the processes in the system Pu – D. aespoeensis (Fig. 2).

The interaction with plutonium includes five processes with different time scales. Interestingly, the physiologically very different soil bacteria Pseudomonas stutzeri and Bacillus sphaericus [3] and D. aespoeensis interact with plutonium in a similar way (processes A to C in Fig. 2). In the first step, Pu(VI) and Pu(IV) polymers are bound to the biomass, e.g.,
phosphate groups of the cell envelope. However, in contrast to the results of Panak and Nitsche [3], who report that one third of the initial Pu(VI) was reduced to Pu(V) after 24h, we observed that 97% of Pu(VI) was reduced to Pu(V) due to the activity of the D. aespoeensis cells within the same period of time. Most of the Pu(V) dissolves back to the aqueous solution due to the weak complexing properties of plutonium in this oxidation state. The dissolved Pu(V) disproportionates to Pu(IV) and Pu(VI). As already described for uranium (Fig. 3), we also assume for plutonium that the structure of the cell membrane is damaged due to metal stress.

Therefore, we conclude a penetration of plutonium species (e.g., Pu(IV)-polymers) inside the bacterial cell from the increased plutonium amount which is strongly bound and not accessible for solvent extractions. It cannot be excluded that additional interaction processes such as the association of Pu(IV) polymeric species with released cell components and/or the uptake of Pu(IV)-polymers by degraded cells take place. Extended X-ray absorption fine structure (EXAFS) measurements of the plutonium accumulated by D. aespoeensis showed that Pu(V) polymeric species are bound to the cells after more than 96 h of contact time. Moreover, an interaction of these Pu(IV) polymeric species with phosphate groups of the biomass is indicated by the isolation of a Pu-P distance at 3.15 Å.

By characterizing the individual processes between D. aespoeensis bacteria and plutonium, this study contributes to a more realistic description of the influence of microbial actions on the migration behavior of plutonium in the environment. What is more, it provides an improved risk assessment for potential underground nuclear waste repositories.

Fig. 2: Illustration of the various processes describing the interaction of plutonium with Desulfovibrio aespoeensis based on the schema developed in [3].

Fig. 3: Transmission electron microscopy (TEM) and energy dispersive X-ray spectroscopy (EDX) analysis of the cellular localization of uranium accumulated by cells of Desulfovibrio aespoeensis.

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Project partners
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Self-organizing surface layer proteins as building blocks for innovative nanomaterials

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Nanobiotechnology is a novel, rapidly growing branch of nanosciences and combines two future technologies: biotechnology and nanotechnology. One important goal of nanobiotechnology is to utilize the unique properties of biomolecules and their interactions for the construction of nanoscale structures and devices. In particular, investigation and utilization of self-organizing biomolecules have attracted much interest in this field.

Surface layer proteins (S-layers), which form the outer sheet around the cells of a large number of primitive microorganisms, are an example of such fascinating self-assembling nanostructures. They form two-dimensional paracrystalline arrays with repeating units on the scale of a few nanometers (10⁻⁹ m). During the last few years, the Institute of Radiochemistry has established their utilization as a technology platform for innovative materials. Among the various S-layer-carrying bacterial strains which have been isolated from uranium mining waste piles, the S-layer of *Bacillus sphaericus* JG-A12 has been extensively characterized, including protein studies, molecular biology, genetic engineering, spectroscopic methods, and its application potential [1, 2]. In particular, it has been used to produce highly regular arrays of inorganic nano-sized grains.

S-layers are ideal templates for synthesis, in situ deposition, or direct patterning of inorganic nanoarrays. Their regularly-distributed pores function as binding sites for various metals such as Pt, Pd, and Au, and offer ideal structures for the formation of regularly-distributed nanoclusters of defined sizes [1]. Such arrays of nanoparticles in a matrix of biological molecules with high stability [2] and defined geometry have great potential for developing novel catalysts, new biomedical and bioanalytical applications, the assembly of nanometer-scaled electronic devices, for the optical industry, and storage media.

Using such materials in technological applications critically depends on the detailed knowledge of their properties on an atomic, nanoscopic, and microscopic scale. In this multidisciplinary field, close cooperation of researchers of various disciplines such as biology, chemistry, physics, and material sciences is essential. In collaboration with the Institute of Radiation Physics, the Dresden High Magnetic Field Laboratory (HLD), and the Institute of Ion-Beam Physics and Materials Research of the FZD, the size, distribution and structure of the nanoparticles, physical properties, and the mechanisms of protein-metal interactions have been analyzed [1–3]. As an example, the properties of S-layer and Pd-nanograin hybrid materials are presented here.

Regarding palladium nanoclusters with mean grain diameters down to 1.5 nm, a clear reduction of the magnetic properties, e.g., of the magnetic susceptibility, compared to the features obtained from bulk palladium has been observed [3]. It appears that ferromagnetic spin fluctuations on a small length and short
time scale, which magnify the magnetic susceptibility of bulk palladium, vanish almost completely in the palladium nano-grains. Additionally, we could demonstrate that iron impurities play a crucial role in enhancing the magnetic properties of the nanograins and qualify earlier findings of ferromagnetism in palladium nanograins with higher impurity concentrations reported by other groups. More precisely, the decrease of the magnetic d-electron band susceptibility of the palladium nanograins corresponds with theoretical models which consider the effect of spin-orbit scattering on the magnetic and superconducting properties of granular, nearly ferromagnetic metals [3]. The weakened magnetism of the d-conduction electrons is considered to play a crucial role for the occurrence of superconductivity in micro and nanogranular platinum by adjusting the balance between electron-phonon interactions and competing magnetic interactions. For this reason, we are now searching also for superconductivity in metal nanoclusters. Also, we have started to investigate nanoclusters of magnetic compounds, such as magnetite, deposited on S-layer proteins as promising hybrid materials for next-generation data storage devices.

For technological applications of S-layers, the stability of protein and nanoparticles under extreme conditions is essential. Unexpectedly, a mutual influence between biological templates and inorganic nanoclusters has been observed. The template effectively influences the growth and spacing of nanoclusters. Vice versa, the inorganic grains stabilize the S-layer. It has been shown that the S-layer of Bacillus sphaericus becomes less sensitive to acidic solutions once it is complexed with PdII+. Usually, proteins irreversibly lose their native structural organization when they are exposed to an acidic environment. S-layers are intrinsically more stable than most soluble proteins. Even more surprising is their gain in stability upon complexation with metal ions, allowing them to withstand very high concentrations of hydrochloric acid without significant loss of their structure. Apparently, the “clogging” of nanopores and crevices on the protein surface by metal ions or metal nanoclusters restricts the motional freedom of the macromolecules in such a way that they are efficiently held together in their native-like structure. Their unfolding into random geometries appears to be inhibited by strong metal protein interactions. By combining infrared and X-ray absorption spectroscopy, it could be shown that mainly the unusually large number of negatively charged carboxyl groups exposed on the S-layer surface engages in binding to the positively charged metal ions [2]. The electrostatic attraction between these coordination partners is the major source of the structural stabilization. This renders metallized S-layers particularly attractive for technical applications where stability at high temperatures is required but rarely met by pure biological materials. The insights gained from metal S-layer interactions may help to confer stability to other technically promising proteins by directed mutagenesis as well.

These findings provide an excellent basis at the FZD for future developments of novel nanomaterials using bacterial self-assembling surface-layer (S-layer) proteins in combination with inorganic nanoparticles as a technology platform.

References
How mineral surfaces retard uranium migration

Vinzenz Brendler, Thuro Arnold, Andreas C. Scheinost, Anke Richter

The migration of uranium from mining areas or nuclear waste disposal areas to ground or surface waters poses a severe threat to both man and nature. Therefore, retention mechanisms to prevent the migration and contamination must be thoroughly understood in order to develop the most efficient and economic countermeasures. The basic processes—interactions between dissolved (here: radioactive) substances and solid phases—range from the weakest form of retention, i.e., ion exchange, to stronger retention mechanisms, such as chemisorption, surface precipitation, and formation of solid solutions. All processes interact in a complex pattern, so that only a detailed understanding of the interactions at the solid/liquid interface allows reliable predictions for uranium migration. The most basic data to collect are the stoichiometry and the structure of the surface species. Such information may be determined only by combining spectroscopy, microscopy, and quantum chemical computations.

Clay rocks are being considered as a possible host rock formation for a future nuclear waste repository in Germany. Investigating the kinetics, thermodynamics, redox behavior, and speciation of uranium in this rock formation often requires comparisons to simpler model systems. Here, gibbsite ($\gamma$-Al(OH)$_3$) or silica gel (SiO$_2$) can mimic interactions between actinides and aluminol or silanol surface groups, whereas kaolinite (Al$_2$Si$_2$O$_5$(OH)$_4$) and muscovite (KAl$_3$Si$_3$O$_10$(OH)$_2$) cover both aspects. In addition, investigations of iron minerals, e.g., ferrihydrite (Fe$_2$O$_3 \times n$H$_2$O), are also highly interesting because iron corrosion products and secondary phases are important components of potential nuclear waste repositories. Most of these mineral phases show a sorption maximum between pH 5 and 8 for uranium(VI). Researchers at the Institute of Radiochemistry determined the basic spectroscopic signal patterns for the uranium binding onto the above model...
systems using mainly TRLFS (Time-Resolved Laser-induced Fluorescence Spectroscopy) and EXAFS (Extended X-ray Absorption Fine Structure). The spectroscopic fingerprints combined with structural information gives access to more complicated systems such as mica, feldspars, or clay rocks.

Gibbsite and silica gel: Within a European Commission ACTINET-6 project with partners from the Institute for Nuclear Waste Disposal at the Forschungszentrum Karlsruhe and the Ecole Nationale Supérieure de Chimie de Paris, we identified three adsorbed U(VI) surface species on Gibbsite by combining TRLFS and EXAFS. These species were a dimer (Fig. 1), a bidentate inner-sphere surface complex, and a uranium carbonate species. The first two species are fluorescent [1], their fluorescence decay times being $330 \pm 115 \text{ ns}$ and $5.6 \pm 1.6 \mu \text{s}$, respectively. The first species dominates the more acidic pH region whereas the second one becomes gradually more prominent towards higher pH values. The fluorescence spectra of both adsorbed uranyl(VI) surface species are red-shifted by 9 nm compared to the free uranyl cation. The spectroscopic fingerprints of these two simplest model systems are distinct enough to allow the assignment of binding sites in more complex systems (such as mica, feldspars, or clay rocks) to aluminol or silanol sites.

Kaolinite: Collaborating with the Institute of Nuclear Chemistry at Johannes-Gutenberg-Universität Mainz, we used EXAFS, XPS (X-ray Photoelectron Spectroscopy) and TRLFS to study the U(VI) surface complexes on kaolinite in presence and absence of humic acid (HA). Two uranyl surface species with fluorescence decay times of $5.9 \pm 0.7$ and $42.5 \pm 1.7 \mu \text{s}$ as well as $4.4 \pm 0.6$ and $30.9 \pm 3.6 \mu \text{s}$ were identified in the binary (U(VI)-kaolinite) and ternary system (U(VI)-HA-kaolinite), respectively. Clearly, aluminol binding sites control the sorption of U(VI) onto kaolinite. In the binary system, both surface species can be attributed to adsorbed bidentate mononuclear surface complexes, which differ in the number of water molecules in their coordination environment. In the ternary system, U(VI) prefers direct binding to kaolinite rather than via humic acid, but it is sorbed as a uranyl-humate complex. Thus, the hydration shell of the U(VI) surface complexes is partly displaced with complexed humic acid, which is distributed between kaolinite particles. Humic acid weakens the retention of uranyl in the neutral pH range, but enforces sorption under more acidic or alkaline conditions.

Muscovite: In cooperation with the Department of Geological Sciences at the University of Michigan (Ann Arbor, USA) we applied TRLFS and HAADF-STEM (High-Angle Annular Dark-Field Scanning Transmission Electron Microscopy) in order to investigate the species of uranyl(VI) adsorbed onto muscovite [2]. HAADF-STEM revealed that nano-clusters of an amorphous uranium phase were attached to the edge-surfaces, but not to (001) cleavage planes of muscovite (Fig. 2), given the main surface site alternatives. TRLFS provided evidence of the presence of two adsorbed uranium(VI) species on these edge-surfaces. The species with the shorter fluorescence lifetimes are interpreted as truly adsorbed bidentate surface complexes, in which the U(VI) binds to the aluminol groups of the edge-surfaces. The surface species with the longer fluorescence lifetimes are interpreted as nano-sized clusters of polynuclear uranyl(VI) surface species with a particle diameter of 1 to 2 nm. These species are intermediate products which are eventually transformed into irreversibly-retarded surface precipitations.
Ferrihydrite: To estimate the mobility of U(VI), we determined the mechanism of U-uptake by fresh precipitates and the molecular structure of U bonding. We employed EXAFS spectroscopy at the Fe K-edge and the U LIII-edge as well as Attenuated Total Reflectance Fourier Transform Infrared (ATR-FTIR) spectroscopy [3]. Freshly formed precipitates were identified as colloidal two-line ferrihydrite. They removed U(VI) from the solution by sorption processes, while surface precipitation or structural incorporation of U was not observed. By employing a novel EXAFS analysis method, the Monte Carlo Target Transformation Factor Analysis, we could ascertain for the first time a 3D configuration of this sorption with a slightly tilted position of the adsorbed UO$_2$$^{2+}$ unit relative to the edge-sharing Fe(O,OH)$_6$ octahedra. In the presence of dissolved carbonate and at pH 8.0, a distal carbonate O-atom at 4.3 Å supports the formation of ternary U(VI)-carbonato surface complexes, which was confirmed by ATR-FTIR. However, in slightly acidic conditions (pH 5–6) in an equilibrium with atmospheric CO$_2$, the U(VI) sorption on ferrihydrite was dominated by the binary complex species -FeO$_2$=UO$_2$, whereas ternary U(VI)-carbonato surface complexes were less relevant. While sulfate and silicate were also present in the mine water, they had no detectable influence on U(VI) surface complexation. Our experiments demonstrate that U(VI) forms stable inner-sphere sorption complexes even in the presence of carbonate and at slightly alkaline pH conditions which have previously been assumed to greatly accelerate the mobility of U(VI) in aqueous environments.

The improved understanding of basic sorption processes, and in particular any independent spectroscopic evidence of surface species that so far has been only postulated, significantly helps to transfer the use of surface complexation models from science into real-world applications. Thermodynamic sorption databases such as RES$^3$T—Rossendorf Expert System for Surface and Sorption Thermodynamics funded by the German Federal Ministry of Economics and Technology (Fig. 3)—and their application toward uncertainty analysis and blind prediction modeling [4] increase the reliability and trustworthiness of the surface complexation models. RES$^3$T is the first and largest mineral-specific sorption database worldwide, currently covering 117 minerals and 130 ligands, providing 3520 surface complexation constants based on 2170 bibliographic references. More than 100 users from 16 countries indicate the high demand for such a tool. The database also offers direct advantages in interpreting reactive transport experiments, as demonstrated together with the Laboratory for Waste Management at the Paul-Scherrer-Institut in Switzerland [5].

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Environmental impact of depleted uranium

Gerhard Geipel, Nils Baumann, Thuro Arnold

The contamination of cultivated areas by uranium in the past is mainly connected to four human activities. Firstly, low-level concentrations of uranium are distributed over large areas by using phosphate fertilizers in agriculture. Secondly, uranium mining and milling has caused uranium contaminations close to residential areas, especially in Germany. The third source of environmental pollution with uranium and radioactive products is connected to nuclear industry by accidents in power stations and processing plants. Last but not least, uranium is used by the military, thus contributing to the distribution of this heavy metal in the environment. Depleted uranium (DU), a byproduct of the enriching process in the production of nuclear fuel, has been used as armor-piercing ammunition in several past wars. Many of these projectiles did not explode completely and residues of the uranium contained in them corrode in the soil. This could be the origin of toxic uranium in drinking water or in agricultural plants. As a consequence, uranium could be transferred into the human food chain. Possible risks for human health have triggered research on the geochemical behavior of DU in nature including transport and immobilization in the soil.

Uranium in its main oxidation states (+4, +5, and +6) shows luminescence properties. Especially in the +6 oxidation state, the stable form under normal environmental conditions, the luminescence of dissolved, solid, or surface species works as a fingerprint to determine the binding forms of uranium [1, 2]. In the past, time-resolved laser-induced fluorescence spectroscopy (TRLFS) has been continually developed. This includes cryogenic techniques as well as the excitation of the species at different wavelengths, leading to extremely low detection limits for the several binding forms. In addition, luminescence datasets that have been collected for most of the uranium binding forms are the basis for fast and correct determination of uranium species.

Uranium ammunition meets dissolved fertilizer

We cooperated with British scientists from the group of Prof. David Read (Enterprise, University of Reading and University of Aberdeen) on a study in which discs of depleted uranium (25 mm diameter, 0.5 mm thickness) obtained from a pristine British military tank shell (Fig. 1) [3] were placed in a low-concentration, well-defined solution of calcium and phosphate and kept there at 24 °C for about half a year. The pH-value of this solution was adjusted to an initial value of 6.0. It gradually decreased to about pH 5.2 by the end of the experiment. The solution concentration of calcium, phosphate, and uranium as well as the pH-value were monitored during the whole experiment. TRLFS measurements on the surface of the sample were carried out and compared with measurements conducted at the same conditions (laser energy, gate width) on an unaltered DU disc, the so-called “blank”, which was not in contact with the Ca-P solution described above. The acquired TRLFS spectra showed six distinct emission bands at 486, 501, 522, 546, and 601 nanometers (Fig. 2), a pattern typical of U(VI). The fluorescence lifetimes were calculated as 50 ± 5 and 700 ± 25 ns.

The shorter lifetime of 50 ± 5 ns was interpreted as a result of the inhomogeneity and disturbances in the crystal lattice on the surface. The observed red shift towards higher wavelengths compared to the spectra of the so-called free uranyl in water (UO$_2$$_{(aq)}^{2+}$) is mostly indicative of uranyl binding forms. As the contact solution contains phosphate ions, the fluorescence spectra of the U(VI) phase on DU were compared with TRLFS data from well-known U-phosphates. Corresponding uranyl phosphate minerals were provided courtesy of the mineral collection of the TU Bergakademie Freiberg [4]. By far the best match among the spectra measured on altered DU was found for the spectra of a natural mineral meta-autunite, Ca(UO$_2$)$_2$(PO$_4$)$_2$ x(H$_2$O)$_8$ (Fig. 2). Here, we found the best agreement in peak maxima, peak height ratios, and in the lifetimes of the fluorescence signals. So far, evidence for the formation and the identification of such a thin mineral layer could only be obtained with the analytical method of TRLFS.
Corrosion of expended uranium ammunition in soil

The experiment described above was able to show the behavior of DU in a solution of a phosphate fertilizer. While this referred to well-defined laboratory conditions, periods of rain and dryness alternate under natural conditions, which can be simulated by lysimeter experiments. At the Institute of Radiation Protection at the GSF-National Research Center for Environment and Health, the corrosion and leaching of depleted uranium (DU) was investigated for three years using six DU munitions (Fig. 3), each buried in a column with a soil core [5]. The first soil was made up of a sandy-loamy brown earth whereas the second soil consisted of a silty-loamy parabrown earth. The columns were installed in an air-conditioned laboratory and were weekly irrigated with 16 mm synthetic rainwater of pH 6.

On average, 7.9% of the initial DU mass was corroded after three years, indicating an acceleration of corrosion compared to the first year. The leaching rates increased stronger than the corrosion by a factor of more than 100, resulting in a mean total amount of leached $^{238}$U of 13 mg as compared to 0.03 mg after the first year. The uranium binding forms present in the seepage water were predominantly hydroxo compounds such as $\text{UO}_2\text{H}_x(\text{OH})_{5-x}$ or carbonate compounds such as $\text{UO}_2(\text{CO}_3)_3^{4-}$ (Fig. 4). Due to evapotranspiration and depending on the position of the ammunition, upward flows of U were observed in all columns, but with different extents. During the second year, in the columns in which the DU was in top position, yellow material crystallized out at the soil surface. Speciation analyses by means of TRLFS revealed that in the yellow material, mainly phosphate species of uranium were present (Fig. 5), probably $\text{AlH(UO}_2\text{PO}_4\times 16(\text{H}_2\text{O})$ (sabugalite) or alternatively $\text{Ca(UO}_2\text{PO}_4\times 10-12(\text{H}_2\text{O})$ (autunite). In the corroded material of the munitions, other phosphate species such as $\text{UO}_2\text{HPO}_4$ were found. Interestingly, the uranium complexes in the seepage water do not seem to be the same as in the corroded mass and the crystallized material at the soil surface.
respectively. It was concluded that the dramatic increase in leaching and its large temporal and spatial variability do not allow any extrapolation into the future. However, the high level of the uranium concentrations in the seepage water requires additional investigation about the transport of uranium through the soil. In so doing, the concentration of uranium from DU munitions in the groundwater in areas affected by DU weapons can be estimated.

There is no other analytical method that could deliver comparable results. It can be concluded that under the circumstances described here, the dissolving process of uranium projectiles is connected to the formation of phosphate-containing secondary minerals. The mobility of DU originating from projectiles is defined by dissolving processes from the emerging secondary mineral phase. With this exemplary investigation, the Institute of Radiochemistry was able to show that interactions between DU ammunition with water and soil, i.e., the reaction mechanism, can be clarified. However, at this stage, predictions can only be made with great caution!

![Naturally accruing meta-autunite sample from the Technische Universität Bergakademie Freiberg mineral collection.](image)

References


Project partners

1University of Reading, United Kingdom
2Institute of Radiation Protection, GSF-National Research Center for Environment and Health, Neuherberg, Germany
3Institute of Mineralogy, Technische Universität Bergakademie Freiberg, Germany
Natural aquatic and terrestrial environments exert wide variations in redox state due to oxygen diffusion and microbial processes. Actinides, with their different oxidation states, are especially susceptible to these redox changes, forming a variety of dissolved complexes which may greatly differ in solubility and mobility. These complexes are often difficult to investigate due to their thermodynamic metastability.

This problem can be solved by using an electrochemical cell, which allows to control and stabilize the redox conditions.

We therefore developed such a cell (Fig. 1) for in-situ X-ray absorption spectroscopy at the Rossendorf Beamline (ESRF, Grenoble, France). The cell comprises a double compartment against radionuclide release, electrodes for electrolysis, and sensors for temperature, pH, and Eh measurements.

Fig. 2 shows the reduction process of uranium(VI) in a formic acid solution. An X-ray absorption near-edge structure (XANES) spectrum was obtained every 30 minutes without interrupting the electrolysis. The stepwise change of the spectral features shows the formation of uranium(IV). A quantitative analysis of the spectra reveals the reduction kinetics of U(VI), indicating that the process is completed after about five hours. While maintaining the redox potential at the final level, extended X-ray absorption fine structure (EXAFS) measurements are then performed to determine the structure of the complexes.
Fig. 3 shows the structure of U(IV) sulfate in aqueous solution as elucidated by EXAFS spectroscopy. The pure U(IV) hydrate is coordinated by nine water molecules with a U-O distance of 2.40 Å. The spectrum of U(IV) sulfate comprises two additional peaks at 3.08 Å and 3.67 Å, indicative of bidentate and monodentate sulfate groups, respectively [2]. The monodentate coordination generally prevails at low [SO₄²⁻]/[U(IV)] ratios, whereas the bidentate coordination becomes more important at higher sulfate concentrations.

The spectroelectrochemical cell is used to study actinides under controlled redox conditions to simulate the geochemical situation in former uranium mines and potential nuclear waste repositories. The development of this new experimental technique was fundamental for several international projects [3-8] funded by the European ACTINET Network of Excellence and the German Research Foundation (DFG) (HE 2297/2-1).

**References**


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**Project partners**

1Centre National de la Recherche Scientifique, Institut de Physique Nucléaire, Orsay, France

2Commissariat à l’Energie Atomique, Marcoule, Bagnols sur Cèze, France

3SUBATECH, Université de Nantes, Ecole des Mines, Nantes, France

4Research Laboratory for Nuclear Reactors, Tokyo Institute of Technology, Japan

5Chemistry Division, Argonne National Laboratory, Argonne, USA

6Department of Chemistry, Katholieke Universiteit Leuven, Belgium
The Forschungszentrum Dresden-Rossendorf (FZD) is a multi-disciplinary research center for natural sciences and technology. It is the largest institute of the Leibniz Association and is equally funded by the Federal Republic of Germany and the Federal States, in particular by the Free State of Saxony. At the FZD, around 225 scientists are engaged in three different research programs of basic and application-oriented research. Scientists working in the Structure of Matter program investigate the reactions of matter when influenced by high fields and minuscule dimensions. Research and development in the Life Sciences program is focused on the imaging of tumors and the effective radiation treatment of cancer. How can humankind and the environment be protected from technical risks? – This question is in the center of research in the Environment and Safety program of the FZD.

In the following Facts & Figures section data presenting the scientific output in the Environment and Safety research program are given as well as information on staff and funding at the FZD.
### Scientific staff

This chart shows the evolution of posts occupied by scientific personnel in the Environment and Safety program of the FZD. Third-party funded scientists, guest scientists, and postdocs represented by the corresponding figures are given in units of paid full-time posts.

### Budget

This table displays the share of each research program as well as the experimental facilities located at the FZD of both public and project funding during the last three years.

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<th>Research Programs</th>
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### Patents

This figure shows the number of applications for a patent filed in each research program of the FZD during the last years. National and international applications for a patent of one and the same invention are only counted once.

### Publications

This chart displays the evolution of peer-reviewed articles by scientists from the FZD’s Environment and Safety program. The figure includes reviewed proceedings (2004: 39, 2005: 50, 2006: 53).
This figure shows the evolution of the doctoral students at the FZD from 2004 until 2006.

Here, the distribution of the international guest scientists who visited the FZD for the purpose of research between 2004 and 2006 is shown according to their countries of origin.
## Organizational Chart

### General Assembly

### Supervisory Board
Chairman: Dr. Gerd Uhlmann  
Deputy: Dr. Jan Grapentin

### Scientific Advisory Board
Chair: Prof. August Schubiger

### Scientific Technical Council
Chair: Prof. Frank-Peter Weiß

### Board of Directors
Science Director  
Prof. Roland Sauerbrey  
Administrative Director  
Dr. Dr. h. c. Peter Joehnk

### Support Staff

### Scientific Advisory Board

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<tr>
<th>Dresden High Magnetic Field Laboratory</th>
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### Life Sciences

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### Environment and Safety

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### Research Technology

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### Institute of Ion Beam Physics and Materials Research

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### Institute of Radiation Physics

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### Institute of Radiochemistry

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### Laser-Particle Acceleration

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