



Institute for
Transuranium Elements



INSTITUTE
ANNUAL REPORT
2004

The mission of ITU is to protect the European citizen against risks associated with the handling and storage of highly radioactive elements. ITU's prime objectives are to serve as a reference centre for basic actinide research, to contribute to an effective safety and safeguards system for the nuclear fuel cycle, and to study technological and medical applications of transuranium elements.



European Commission

Joint Research Centre

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CONTENT

FOREWORD 3

OVERVIEW Scientific Objectives 4
Events 5
Quality Management 7
Facts & Figures 8

ACTIONS

1. Basic Actinide Research 10

HIGHLIGHT • *Single Crystal Growth of "115" Actinide Compounds*
REVIEW ARTICLE • *Operation of the Actinide User Laboratory at the ITU: Transnational Access to Research Infrastructures Programme 2002-2004*

- 1.1 Solid State Physics of Actinides and Compounds
- 1.2 Electronic Structure of the Actinide Hydrides (AnH_3): A Photoemission Study

2. Spent Fuel Characterisation 24

HIGHLIGHT • *Limits of the Radiolytic Enhancement of Spent Fuel Dissolution in Groundwater: Activity Dependence*

- 2.1 Redox Conditions, Hydrogen Effect and Radiolysis
- 2.2 Studies on the Corrosion Behaviour of Pu-containing Fuel
- 2.3 Effect of Alpha-decay on Long-term Behaviour of Irradiated Fuel

3. Safety of Nuclear Fuels 34

HIGHLIGHT • *Thermophysical Characterisation of UO_2 Fuel with a Burn-up up to 100 MWd/kgHM*

- 3.1 Commissioning Tests on a Shielded Secondary Ion Mass Spectrometer for the Analysis of Irradiated Nuclear Fuel
- 3.2 HTR Fuel Testing
- 3.3 Development of a Method for the Production of Inert Matrix Fuel Kernels for Plutonium Disposition in High Temperature Reactors (HTRs)
- 3.4 Matrix Swelling Rate and Cavity Volume Balance of UO_2 Fuels at High Burn-up
- 3.5 Further Development of the TRANSURANUS Code
- 3.6 PHEBUS PF Post Irradiation Examination (PIE) of the Degraded FPT2 Bundle

4. Partitioning and Transmutation 44

HIGHLIGHT • *First Americium Fuel Fabrication in the Minor Actinide Laboratory*

- 4.1 Partitioning
- 4.2 Studies of Advanced Nuclear Processes
- 4.3 Investigation of the Thermodynamic and Vibrational Properties of Ceramics for the Immobilisation of Actinides

5. Alpha-Immunotherapy / Radiobiology 50

HIGHLIGHT • *From Bench to Bedside: Targeted Alpha Therapy (TAT) for Malignant Melanoma*

- 5.1 Production of Radionuclides for Targeted Alpha Therapy: ^{225}Ac / ^{213}Bi
- 5.2 Pre-clinical and Clinical Studies
- 5.3 Radiobiology

6. Measurement of Radioactivity in the Environment 55

HIGHLIGHT • *Characterisation of Environmental U- and Pu-rich Particles by Synchrotron Radiation Based Techniques*

- 6.1 Measurements of Radioactivity in Bulk Samples
- 6.2 Environmental Radioactive Microparticles

7. Safeguards Research and Development 61

HIGHLIGHT • *Characterisation of Radioactive Microparticles for Nuclear Safeguards Verifications and Detection of Undeclared Nuclear Activities*

- 7.1 Improvement of Measurement Techniques and Methods
- 7.2 Detection Methods for Measurement of Microparticles
- 7.3 Analytical Support
- 7.4 Illicit Trafficking and Nuclear Forensic Science
- 7.5 Operation of the Laboratoria In Situ
- 7.6 Support to the IAEA

ANNEX Contributors 73

ABSTRACT 75

Enlargement in May 2004 was something for which ITU worked hard for several years. Workshops and visits in 2004 were held bringing more than 200 scientists and specialists in a wide range of disciplines to ITU. Our personnel also visited the new member states, participating in workshops, and organizing exercises on detection of illicit trafficking at border crossing in Lithuania, Poland, and Slovakia in 2004. Many such exercises and workshops were held in other countries in previous years and have been discussed in earlier reports. Although a few days before the actual „day“ – 1 May 2004 – the new flag poles appeared to resemble masts in a wind swept harbour; on the actual day they were all wonderfully vertical and a slight breeze made a spectacular view with the European and 25 member state flags flying proud from the masts. Many friends from the new countries joined us for a fine party that opened with the Beethoven's 9th symphony and finished with speeches and champagne.



Gerard Lander
Director

Our „core business“ remains science, and in 2004 there was much of it. No less than 6 workshops this past year gave a sense of the breadth and excitement of the scientific profile of the Institute. These are discussed in the section on „events“ with suitable photos, but the collaborations established and the ideas exchanged remain woven into the fabric of ITU.

On 6-July after an extensive qualification phase, the Minor Actinide Laboratory went into active operation. A batch of americium oxide was introduced in the boxes, for the fabrication of pellets and pins for the CAMIX/COCHIX project. Well done to all the team!

We also had our share of visitors, notably visits from Hans-Gert Pöttering, presently chairman of the European People's Party at the European Parliament, accompanied by a number of other MEP's and from Wolfram König, President of the German Bundesamt für Strahlenschutz to discuss a series of collaborations, primarily in the area of detecting and stopping illicit trafficking of nuclear material.

At the end of 2004 there were 32 grantholders (students and postdocs) resident at the Institute, almost 50% of them female. This is the largest number of grantholders ever at ITU, and it's a wonderful sign for the future of science and the Institute.

My sincere thanks to all the technical and support staff for a year of considerable progress; in particular, to the Management Support Unit for their efforts to interpret and overcome the many new rules imposed by the Commission after the reform.

Scientific Objectives

A brief description of the objectives of the ITU research programme is given below:

Spent Fuel Management

The safe disposal of highly active waste and spent nuclear fuel dominates the nuclear debate in several countries. The two approaches for spent nuclear fuel management favoured by the Member States consist of intermediate storage with subsequent conditioning for final disposal, and intermediate storage with subsequent reprocessing (partitioning) for lowering the radiotoxicity (transmutation) before final disposal in geological formations.

- The work performed at ITU aims at discerning the mechanisms of leaching of irradiated uranium oxide and MOX fuel in water, studying the solid state physical-chemical properties of irradiated fuel, and comparing the results of theoretical models.
- Partitioning and transmutation research should help at further reducing the potential long-term hazard of wastes. The objective is to efficiently separate the long-lived nuclides from the waste, to develop the technologies to allow their incorporation in well-designed fuels or targets, and to transmute them in dedicated reactors by neutron capture or fission into nuclides with much shorter half-lives. A novel line of research is possible transmutation using lasers. The conditioning of actinides in ceramic matrices is also studied.

Safety of Nuclear Fuel

The safety of nuclear installations remains a major public concern even if energy production by nuclear fission is considered a mature technology. Licensing authorities request safety improvements and industry tries to increase operational efficiency. The understanding of the behaviour of nuclear fuel under incident and accident conditions allows improvement of the response and precautions to be taken if such events occur. The experimental investigations also deliver input to the TRANSURANUS fuel performance code. The objective of ITU is to contribute to nuclear safety improvements by studying in detail the behaviour of fuel rods and their integrity in light water reactor at extended times.

Safeguards R&D

Stopping the further spread of nuclear weapons is a major policy objective of the European Union. The inspection authorities, DG TREN (Transport and Energy) of the European Commission and the International Atomic Energy Agency, are charged with the implementation of

safeguards measures to control the use of nuclear materials within the European Union and worldwide. The Institute is a long-standing partner of the Safeguards Inspectorates providing assistance and expertise particularly relevant to plutonium handling facilities, such as reprocessing and MOX fuel fabrication plants. For more than 3 decades, ITU has been providing new analytical and characterisation tools. Since 1999, it operates and maintains on-site laboratories in the reprocessing plants of la Hague and Sellafield.

Radioactivity in the Environment

The protection of citizen and environment against ionizing radiation requires accurate and precise analytical methods for the detection and quantification of radionuclides in the different ecosystems. Moreover, there is a stringent need for the independent verification of radioactive releases into the environment. The JRC competence in trace detection and analysis is used to develop projects for the surveillance of radioactivity in the environment, including studies on migration patterns of actinides in the biosphere and assistance in radiological emergencies.

Basic Actinide Research

A good knowledge of fundamental physical, chemical and material science data on actinides is a pre-requisite for solving any of the above mentioned problems. Basic actinide research aims at advancing the basic knowledge necessary in support to EU policies dealing with nuclear fuels, nuclear waste or nuclear safeguards. In the frame of this action, ITU scientists aim at developing a detailed understanding of actinides and actinide compounds, including their electronic structure, equation of state and phase diagrams, solution chemistry, interface and surface phenomena. This is complemented by various training programmes, in order to maintain in the domain a high level of scientific competence for Europe.

Alpha-Immunotherapy and Cell Toxicity

Cancer continues to be one of the major preoccupations of the European citizen and one of the great challenges for research. ITU supports in cooperation with university hospitals, the development of a new type of cancer therapy based on the use of alpha-particle emitting nuclides. The objective of ITU is to make its facilities and expertise in handling highly radioactive alpha-nuclides available to develop, test, validate and optimise a drug against different types of cancers.

Events

1st Technical Meeting on X-ray Tomography of Nuclear Materials

JANUARY • In collaboration with Forschungszentrum Karlsruhe and the ESRF (European Synchrotron Radiation Facility, Grenoble), ITU organised the 1st Technical Meeting on X-ray Tomography of Nuclear Materials. The first applications of high-resolution X-ray tomography, based on the use of synchrotron radiation, to the study of nuclear materials were presented. The main advantage of tomography with respect to other techniques is that it enables a 3D image of the material microstructure to be obtained. About 30 international experts agreed on the necessity to extend the studies to more active samples.

Journées des Actinides (JdA)

APRIL • The “Journées des Actinides” (JdA) is an annual conference for European and international scientists involved in research programmes on the physics and chemistry of the 5f elements (actinides). The “34iemes Journées des Actinides”, the largest meeting ever, was organised by ITU and held in Heidelberg with 116 participants coming from 17 different countries.

There were lively debates with oral presentations and posters, and especially significant was the round table discussion held on the future of the Actinide User Laboratory at ITU.

Prior to the JdA, a school on the “Physics and Chemistry of Actinides” was held at ITU. The School consists of invited experts giving elementary-level lectures on a wide variety of physics and chemistry in actinide science that are of present concern to the actinide community and was attended by 35 students.



David Bottomley explains the Hot Cells to the participants of the workshop

Radioactivity, Radionuclides & Radiation Training Courses

APRIL AND OCTOBER • The 3rd and 4th edition of the *Radioactivity – Radionuclides – Radiation* training courses with Nuclides.net consisted in lectures and case studies given by ITU staff on actual projects underway in ITU and ranged from the “age” of uranium materials, to laser transmutation experiments and the transport of radioactive materials.



Nuclides.net training course

Workshop on the High Burn-up Structure

JUNE • About 30 international specialists discussed the R&D efforts in the field of the high burn-up structure formation and its consequences on the behaviour of nuclear fuels. This structure appears in high burn-up fuels and corresponds to a new morphology for the material with different thermal properties as compared to fresh fuel. R&D activities have been launched world-wide in order to characterize this structure and to assess its impact on fuel performance. Experimental results and an understanding of the fission gas release and swelling in the high burn-up fuels were discussed. The needs for implementation of new models in the fuel performance codes were also examined.



HBS workshop participants

4th Alpha-Immunotherapy Symposium

JUNE • ITU, in co-operation with the Düsseldorf University and the German Cancer Research Centre, organised the 4th Alpha-Immunotherapy Symposium in Düsseldorf. It allowed more than 70 experts to discuss fundamental and applied aspects of alpha-immunotherapy. The clinical applications of targeted alpha-therapy for leukaemia, lymphoma and malignant melanoma concretized in phase I and II studies, which are supported by ITU with the production of $^{225}\text{Ac}/^{213}\text{Bi}$ generators, were discussed in detail, as well as the potential for new applications. Other topics were isotope production and radiochemistry, dosimetry, radiobiology and preclinical models for alpha-immunotherapy. Currently, a total of 10 different cancer types are under study in pre-clinical and clinical approaches. A new application is the use of alpha-immunotherapy for fungal and bacterial infections.



Schloss Jägerhof, Düsseldorf

Lasers & Nuclei – Applications of Ultra-high Intensity Lasers in Nuclear Science

SEPTEMBER • The prediction of nuclear fission induced by laser light was made in 1988 and demonstrated experimentally in 2000 through collaboration with UK universities, Rutherford Appleton Laboratory in UK, and ITU. Through this development, nuclear reactions can now be induced and studied in the laboratory without recourse to nuclear reactors or particle accelerators. As a result, a new field of Laser Nuclear Physics has emerged and is currently undergoing tremendous growth. International experts and representatives from leading groups discussed together the applications in nuclear and astrophysics, medical applications, transmutation,

radiography and addressed the questions of how these lasers could make a direct impact on technology. The final session described future developments in high intensity lasers.



Lasers & Nuclei workshop

11th Symposium on the Thermodynamics of Nuclear Materials

SEPTEMBER • At the 11th Symposium on Thermodynamics of Nuclear Materials the latest results and the current trends were presented focusing in thermodynamics of oxide fuel materials and actinide materials. With an increasing number of modelling studies, and a decreasing number of experimental studies, attention concentrated on Pyrochemistry and Advanced Fuels field due to their increasing importance for the nuclear fuel cycle. The Environment and Waste session focussed on long-term stability of spent fuel and waste forms, where thermodynamics, as well as kinetics, must be taken into account. Much discussion took place between the participants and many networks and potential collaborations developed during the meeting.



Schloss Karlsruhe

Quality Management

Considerable efforts for integration and simplification of ITU's management system were undertaken to foster further development and user friendliness following the implementation of new version of the software tool QM Intra.

All internal processes were reviewed and QM Intra updated accordingly and expanded to include further details about internal processes, links to applying regulations and information concerning management and handling of dangerous substances, management of transport of radioactive materials and disposal of radioactive materials. More procedures and working instructions have now been directly linked in electronic format for easy access. QM Intra is thereby contributing to the documentation and preservation of the Institute's know-how and knowledge.

Our certificate ISO 9001:2000 was renewed through DQS and is now valid until April 2007. Recommendations of DQS auditors are analysed and checked for applicability prior to implementation. They contribute to understand the interactions of and to help to develop our management system.

The Co-ordination Group decided to do a follow-up of the self-assessment (SA) of the previous year rather than preparing a new SA in 2004. This approach was chosen to make use of the information gathered during the 2003 exercise. A self-assessment follow-up workshop was therefore organised in July during which Heads of Units and members of the SA team of 2003 discussed and interpreted results of SA 2003 trying to maximise the benefit of the exercise namely in areas such as knowledge management, staff management and project management.

The preparation for the accreditation according ISO 17025 of the Analytical Services (AS) of Nuclear Chemistry concerned:

- Training of staff of the AS, also seven ITU staff members were selected and trained as internal auditors
- The internal audit system was reviewed to include audits according to this standard and two internal audits were conducted within AS
- Systems, documents and procedures were reviewed and included into QM Intra
- An accreditation body was selected

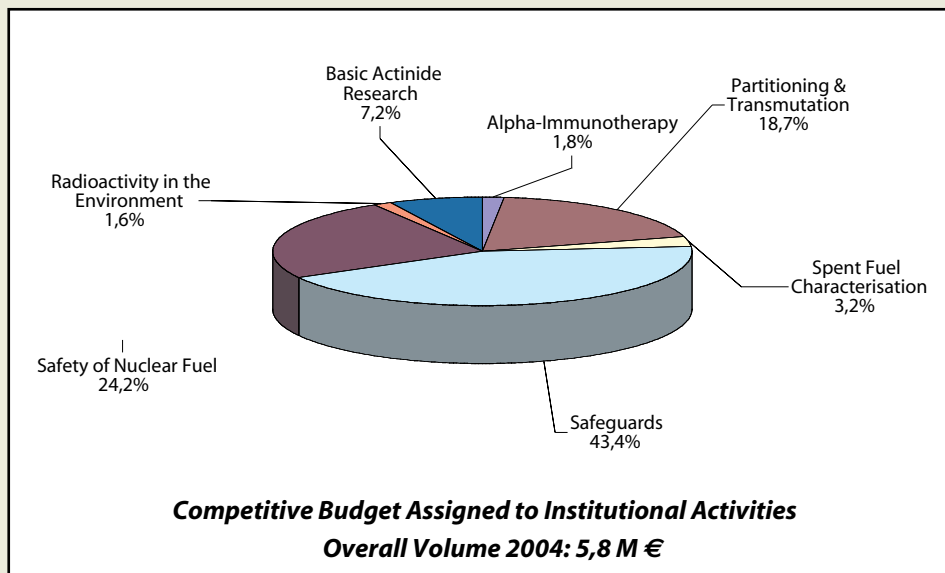
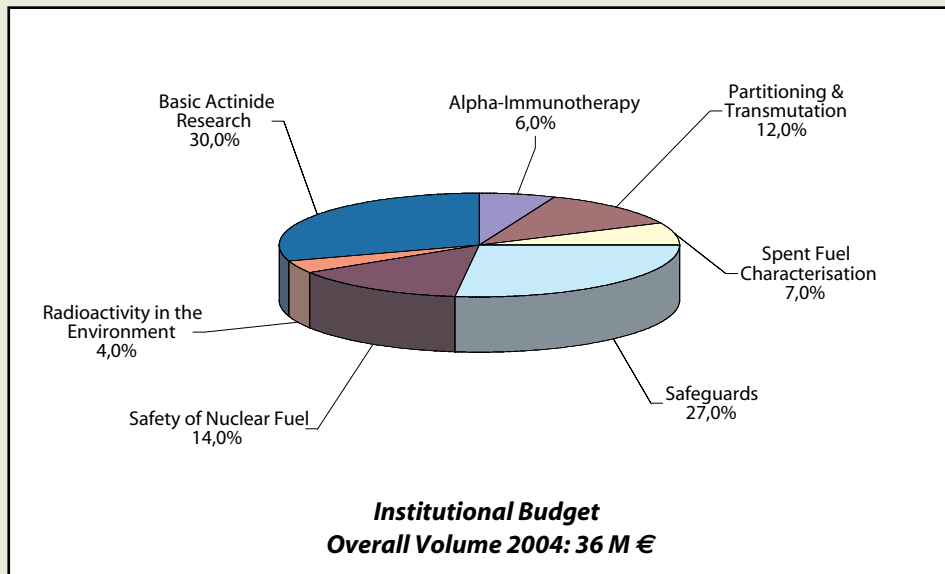


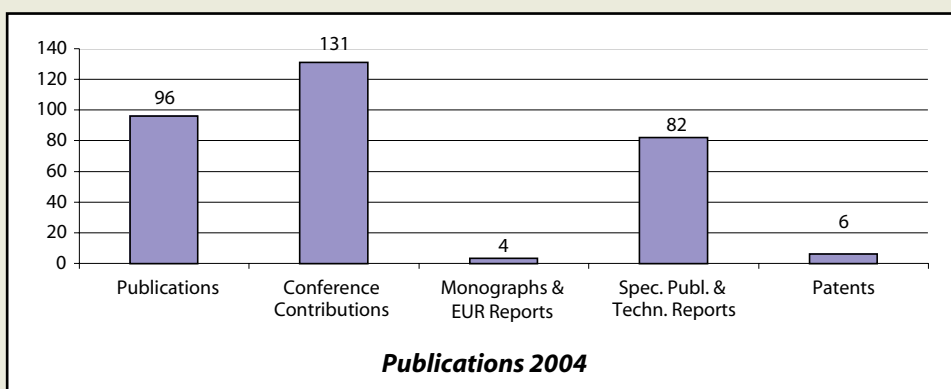
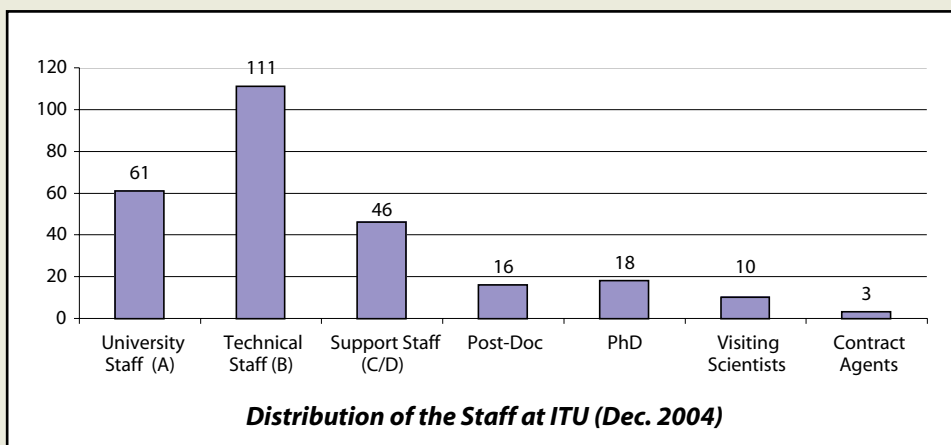
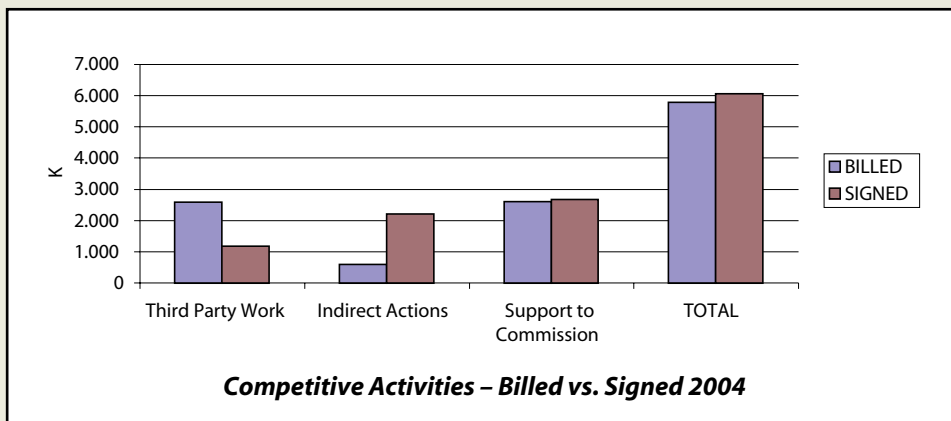
Omer Cromboom and Karin Casteleyn proudly showing the certificate of accreditation of Analytical Services

Finally in late November the audit for the accreditation took place through DACH*. All fifteen techniques that were examined passed the audit. For the Analytical Service this was a very important achievement. It allows them to continue contributing to and being part of international networks e.g. of the International Atomic Energy Agency (IAEA).

* DACH: Deutsche Akkreditierungsstelle Chemie

Facts & Figures



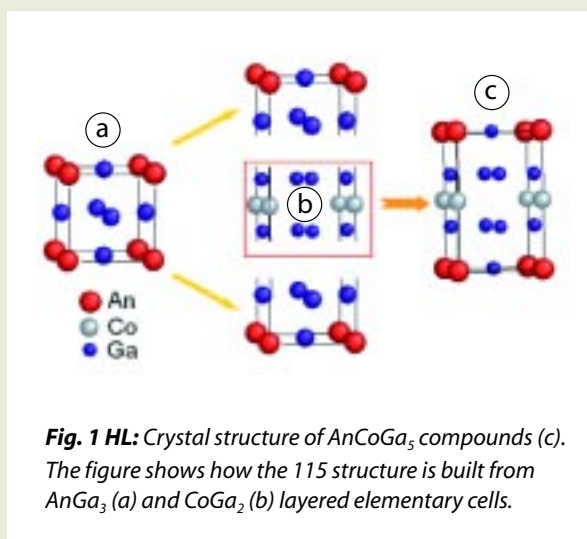


HIGHLIGHT

Single Crystal Growth of "115" Actinide Compounds

The discovery of superconductivity at the surprisingly high temperature of 18 K in the compound PuCoGa_5 [1] has greatly increased the interest of the physics community in such compounds. Since the highest superconductivity in uranium compounds is ~ 2 K, this order of magnitude increase in the superconductivity in a plutonium compound is especially intriguing in the sense that the form of superconductivity might well be different when passing from uranium to plutonium. Theorists have been especially active, with a number of ideas already advanced.

Many experiments have already been performed on polycrystalline samples of these 115-type materials, but the information content from such experiments is limited, especially if extra phases are present. To understand the microscopic origins of the superconductivity special samples in the form of single crystals are required. The growth of crystals will automatically involve cleaning of the starting material due to segregation of impurities during the growth process. For the anisotropy of the superconducting properties we may benefit from the large amount of work presently being done worldwide on high T_c materials; e.g. on the material $\text{YBa}_2\text{Cu}_3\text{O}_7$. Such high T_c materials, in which the superconducting transition temperature now exceeds 100 K, have two-dimensional properties in that the superconducting electrons are confined to the Cu–O planes in the material. The crystal structure of the PuCoGa_5 is shown



in Fig. 1 HL and there are clearly planes with Pu–Ga atoms. Do they “carry” the superconductivity?

Suitable experiments to establish this have not yet been done, although some theories have already predicted such anisotropy in the properties. Thus, further progress in the Pu 115 compound studies depends critically on the availability of single crystals.

Actinide single crystal growth is one of the core competences of the Institute for Transuranium Elements. Since the early 80s several crystal growth techniques have been developed and applied to actinide systems [2]. In the case of the 115 compounds, the method developed at ITU is the same as used at other places and depends on precipitation out of a gallium flux. This has been most successful and single crystals of PuCoGa_5 even with the non-fissile ^{242}Pu isotope have been made, see Fig. 2 HL. The use of the ^{242}Pu isotope, which has a much lower activity than ^{239}Pu , means that the samples with ^{242}Pu may be cooled to lower temperatures than those with ^{239}Pu , and, equally importantly, neutron experiments can be performed whereas the slow neutron fission cross section of ^{239}Pu is too high. Moreover, the method was spectacularly successful in the case of AmCoGa_5 , where a crystal of approaching 1 cm^3 was made as shown in Fig. 3 HL.

Another example of the anisotropy of these compounds can be found in their magnetic properties. One of the “favourite” interactions that could cause the formation of Cooper pairs in the Pu-115 materials is a “magnetic interaction” via spin fluctuations, analogous to phonons causing the pairing in normal BCS-type materials. Thus, it is impor-

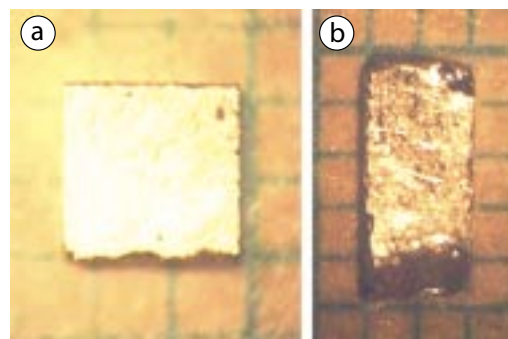


Fig. 2 HL: Photographs of PuCoGa_5 single crystals isolated from the Ga-flux: (a) a cube shaped crystal for anisotropy measurements and (b) a 166 mg $^{242}\text{PuCoGa}_5$ single crystal.

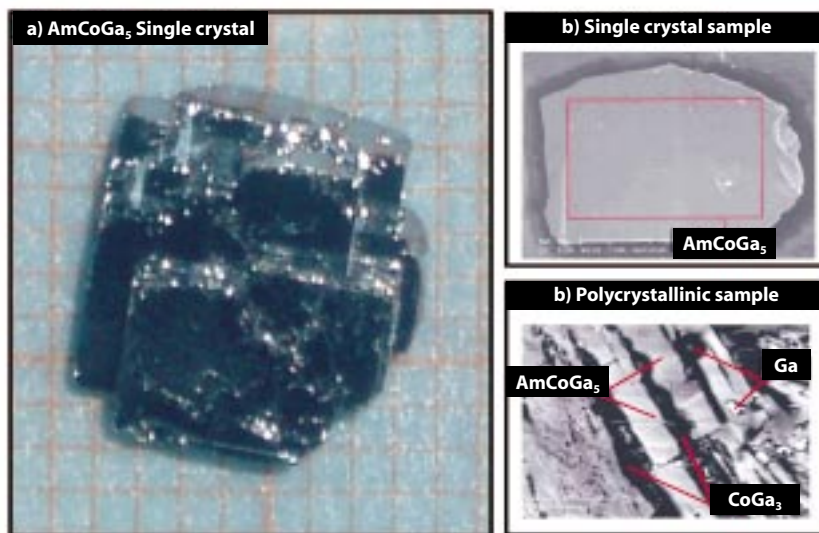


Fig. 3 HL: Photograph of AmCoGa_5 large single crystal (a) as obtained by Ga-flux growth. For comparison, microstructure analysis of a single crystal (b) and a polycrystalline (c) samples are shown.

tant to study and understand the magnetic interactions in these materials so that a complete characterization of their electronic properties may be obtained. The Np-115 materials are *not* superconducting, but are magnetic. Fig. 4 HL shows an example of the magnetization as measured on samples of NpCoGa_5 . The results with the polycrystalline sample show an important transition at 4.5 T, but they do not allow a clear statement of the nature of this transition.

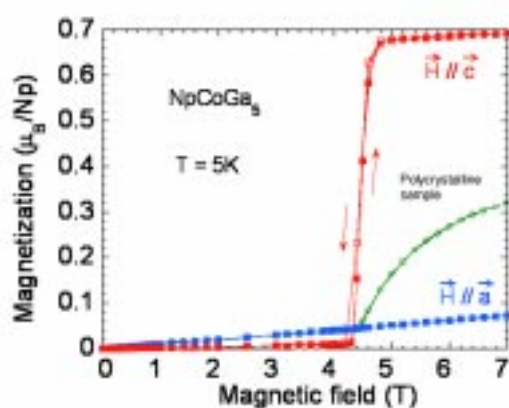


Fig. 4 HL: Anisotropic magnetic behavior of NpCoGa_5 single crystal. The figure shows the magnetization of an oriented NpCoGa_5 single crystal at $T = 5\text{ K}$ with $\vec{H} \parallel \vec{c}$ and $\vec{H} \parallel \vec{a}$. The magnetization of the polycrystalline sample is also shown, for comparison.

As seen from the results from the single crystal sample, the effect *only* occurs when the magnetic field is applied along the c -axis and there is *no* effect when the field is perpendicular to this axis, at least up to the fields available in the laboratory (7 T). In addition, the value of the magnetization when the field is applied in the easy direction ($\vec{H} \parallel \vec{c}$) is $0.70 \mu_B$, a value more than twice as great as that deduced from the polycrystalline sample. Since this value is a feature of the ground-state properties of NpCoGa_5 , the importance of using single crystal samples is evident.

Many new experiments are envisaged with the single crystal samples. Their production has opened a new chapter in the work on ITU on this exciting scientific problem.

References

- [1] J. Sarrao et al., Nature 420 (2002) 297
- [2] J.C. Spirlet and O. Vogt, Handbook on the Physics and Chemistry of the Actinides, eds A.J. Freeman and G.H. Lander, Vol. 1, Chap. 2, Elsevier (1984)

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1. Basic Actinide Research

REVIEW ARTICLE

Operation of the Actinide User Laboratory at the ITU: A Transnational Access to Research Infrastructures Programme 2002-2004

The Actinide User Laboratory is part of the Institute for Transuranium Elements. It offers access to performing basic and applied research on actinide materials such as uranium, neptunium, plutonium etc., to external users who do not have dedicated facilities at their home organisation.

Materials preparation facilities and a suite of instruments (single crystal growth, x-ray diffraction at room and high temperatures as well as in pressure cells up to 50 GPa, physical property measurement facilities from room temperature down to 0.3 K as well as in magnetic fields up to 9T, photoelectron spectroscopy, ...) together with expert technical assistance, are available for conducting basic or applied research studies. It addresses a large number of questions related to actinides, both basic and applied, and is the *only* non-classified Laboratory offering access to external users in Europe where research on appreciable quantities of transuranium materials is conducted.

From 2002 to 2004, this Actinide User Laboratory project has been run within the frame of the European Commission programme "Transnational Access to Research Infrastructures" under the contract N° HPRI-CT-2001-00118. About 331 operating days have been delivered to 35 users to perform 34 projects. It is continuing in 2005 on specific credits of the Basic Actinide Research action.

Why an Actinide User Laboratory?

In spite of actinide research being an enormously exciting intellectual challenge, the number of researchers on actinides is decreasing. This can be seen from different perspectives. Nuclear matters are no longer fashionable. Moreover, the number of students attracted by scientific career decreases and they hesitate to enter a field associated with an energy source that is either being abandoned in many countries or does not have a good press. Many universities no longer teach subjects such as radiochemistry, and enrolment in such subjects as nuclear en-

gineering has declined steadily over the last decade. Increased safety restrictions have now made it impossible in many universities to even handle uranium, let alone any transuranium nuclides.

These events must be seen against a backdrop in which the long-term issues posed by the nuclear industry, both as a major source of energy as well as a producer of radioactive waste, must be addressed. Knowledge of the field of actinides is thus vital for the future. Whether one believes that nuclear energy will make a come back when the real consequences of global warming are appreciated, or whether alternative sources can be found for the increasing demand for electricity, especially in the developing world, we must have people who are *informed* about the nuclear options. Independent of the future of nuclear energy, the legacy of the waste, both civilian and military, will be with us for many generations to come.



Review Article Fig. 1: Logo of the Actinide User Laboratory at ITU-Karlsruhe

The expertise and uniqueness of its equipment make the Institute play a central role in actinide research. Its transnational character, positioning the ITU as the European reference centre in the field, makes it the place to address the concern of tomorrow's expert in nuclear fields. Through the Actinide User Laboratory, our aim is to widen the use of our facilities. Researchers, and students performing their theses work primarily on uranium based compounds, in universities or other laboratories, can visit the Institute for short periods to perform either more experiments on their U-based materials or complement their work on analogous or isostructural Np- or Pu-based materials.

The challenge and management issues

The Actinide User Laboratory is by definition installed in a nuclear infrastructure. The first challenge encountered in this project was to define access procedures that could allow a wide range of European scientists to perform short term projects. These procedures involve both security (clearance procedures to nuclear infrastructure) and safety (medical filing and training). This was the first major programme of this sort applied to an infrastructure offering direct access to high risk materials.

The second challenge addressed the coordination of the user projects to be performed within tight time schedule. Some projects required the organisation of transport of nuclear materials (samples prepared in other facilities and transferred to ITU for measurements) that are often delayed due to lengthy procedures.

The third challenge has been to develop a spirit of "user" service within the laboratory and combine both institutional research projects with users training and allocation of operating days (and related manpower) to outside driven experiments.

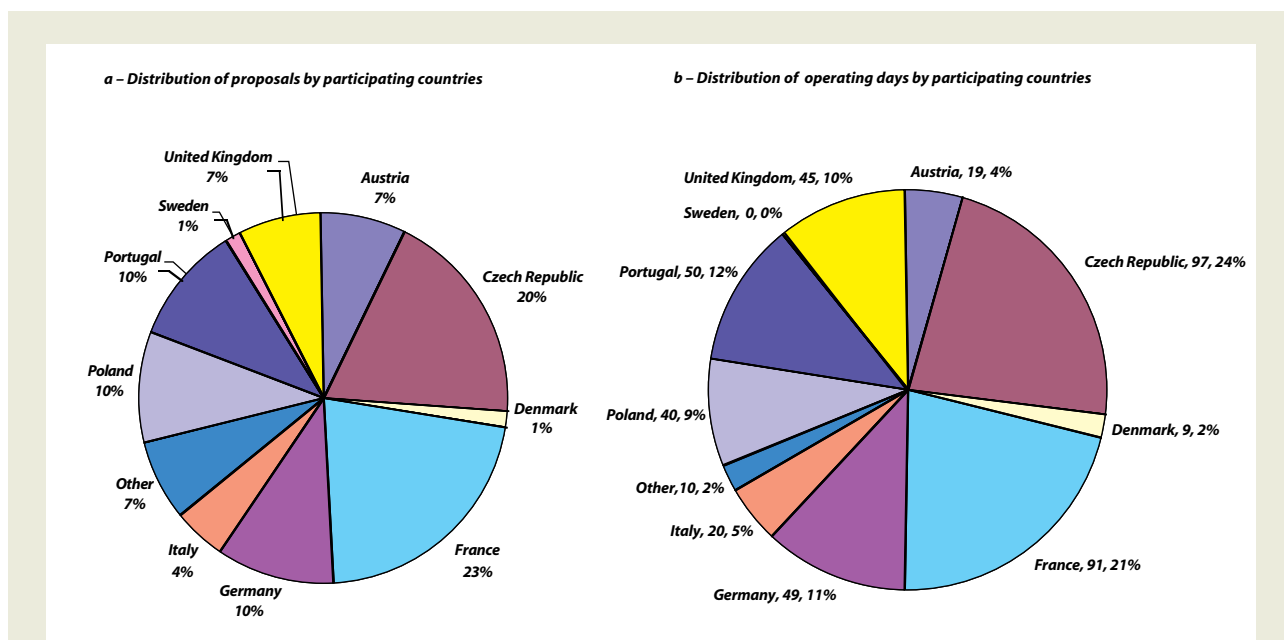
All these challenges were overcome thanks to the engagement of the JRC and ITU personnel from Management Support and Infrastructures Units, and to technical and scientific staff in the laboratory.

Selection procedure and participation

Selection of a user project is made through bi-annual calls for proposals displayed on our website and announcements addressed personally to about 500 potential users listed in a contact database.

The proposals received are reviewed for technical feasibility and assessed by a Selection Panel composed of 4 external independent experts and 2 ITU staff members. Each proposal is scored according to four criteria: *Scientific merit and quality of the proposal*, *Technical feasibility*, *Community interest in the proposal* and *Priority according to educational duty* (i.e. Ph-D student involved, transuranium materials, new users, region, gender balance). As highlighted, the last criteria takes into account "educational" aspects including new users, regions having little access to similar infrastructures, gives emphasis to transuranium materials, and students involvement as well as "gender balance" and proposals of equal scientific merit and interest are set different priority accordingly.

From 2003 to 2004, five calls for proposals were issued and 69 project proposals received. All proposals were assessed and 43 were granted operating days, of which some will be performed in 2005. Figure 2 shows the proposal distribution and allocated operating days by participating countries.



Review Article Fig. 2: Proposal distribution (a) and granted projects (b) by participating countries

1. Basic Actinide Research

Success stories

As discussed above a large number of projects have been performed and we can not describe all of them in detail. However, one can distinguish several classes of projects and illustrate each through selected examples involving different types of the facilities available.

The first and largest class concerns projects that address exploratory research on new type/family of materials. The main interests are centred along the search for new systems showing correlated electron phenomena, new magnetic interactions and interrelation between magnetism and superconductivity. This type of research is widely addressed in rare-earth and uranium systems, whereas more rarely conducted on transuranium compounds (due to the lack of facilities able to handle appreciable quantities of materials).

Crystallochemistry, magnetic, electrical and thermal properties of NpPd₂Sn

Rare-earth compounds $REPd_2M$, where $M = \text{In, Sn or Pb}$, are of interest because of their magnetic and superconducting behaviour [1] with the most fascinating coexistence of both phenomena established for ErPd_2Sn [2] and YbPd_2Sn [3]. In turn, the uranium-based phases UPd_2M are known to be moderately-enhanced heavy fermion systems with the examples being the stannide UPd_2Sn (paramagnetic down to 0.3 K, $\gamma \approx 130 \text{ mJ/molK}^2$) [4] and the plumbide UPd_2Pb ($T_N = 35 \text{ K}$, $\gamma \approx 100 \text{ mJ/molK}^2$) [5]. Motivated by these findings a PhD on the actinide

isostructural compounds started at the Institute of Low Temperature and Structure Research, Polish Academy of Sciences, Wroclaw, Poland. The new NpPd_2Sn compound was synthesised at ITU and shown to be isostructural with its uranium-based counterpart UPd_2Sn . Magnetisation, specific heat (see Fig. 3) and resistivity measurements results are consistent with the scenario of strong electronic correlations.

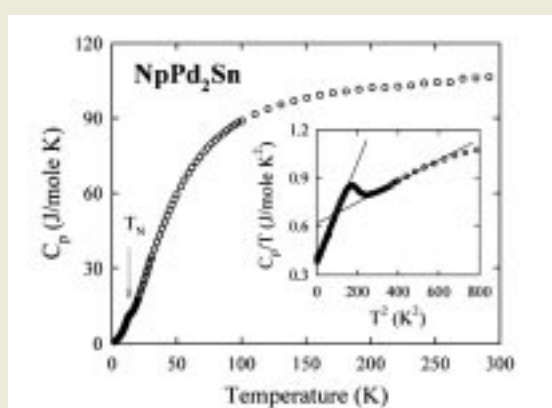
Hence, NpPd_2Sn might be a novel heavy-fermion system (with a Sommerfeld specific heat coefficient γ as large as about 400 mJ/mol.K^2), one of very few known so far amongst neptunium-based intermetallics.

Magnetism of nanocrystalline UN

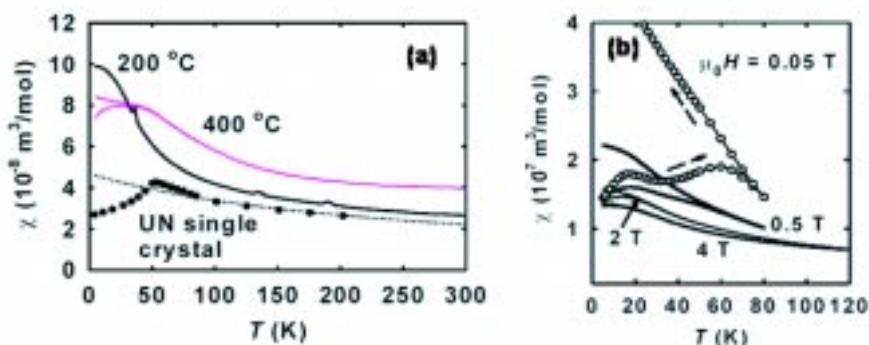
Investigation of nanocrystalline or amorphous new materials is another type of exploratory approach to address new phenomena in magnetism. They have been studied so far mainly in cases relevant for technical applications, i.e. in ferromagnets based on $3d$ or $4f$ elements. The $5f$ magnetism of light actinides, which can be characterised as narrow-band magnetism with large orbital moments, was not investigated systematically yet. As to fundamental interest, the insight in the $5f$ magnetism obtained by the possibility of variable disorder is rather intriguing. A team from Charles University, Prague, addressed this challenge on UN nanocrystalline material. The preliminary results show a dramatic change in the magnetic properties of the $5f$ systems when nanomaterials are investigated. Magnetic susceptibility of UN is normally characterized by a pronounced maximum at $T_N = 53 \text{ K}$ (see Fig. 4a).

In the paramagnetic range, the magnetic susceptibility is described by a large negative $\theta_p = -247 \text{ K}$ and a reduced effective moment $2.66 \mu_B$ [6]. Sputter deposited UN layers do not exhibit the anomaly around 50 K. Instead, $\chi(T)$ increases monotonously with decreasing T . A comparison of $\chi(T)$ in various fields indicates a small ferromagnetic component developing gradually below $T \approx 100 \text{ K}$ (see Fig. 4b).

Generally, these differences can be attributed to not fully compensated antiferromagnetic coupling at grain boundaries (the grain boundaries represent a non-negligible volume fraction) and/or at numerous defects. This situation affects the susceptibility even in the paramagnetic state by contributing an extra term with a small μ_{eff} but $\theta_p > 0 \text{ K}$. Magnetic history phenomena [difference of field-cooled (FC) and zero-field-cooled (ZFC) regimes] show that the blocking of the weakly ferromagnetic clus-



Review Article Fig. 3: Temperature dependence of the specific heat of NpPd_2Sn . The inset shows the low-temperature data in the form C/T versus T^2 . The solid lines mark a straight line behaviour.



Review Article Fig. 4: (a) Temperature dependencies of magnetic susceptibility of the UN layers synthesized at $T_s = 200^\circ\text{C}$ and 400°C compared with the bulk data, taken from the reference given in the text. (b) Temperature dependence of magnetic susceptibility of UN synthesized at $T = 300^\circ\text{C}$. The arrows indicate ZFC and FC regimes for the measurement in $\mu_0 H = 0.05 \text{ T}$. Increasing magnetic field reduces the FC-ZFC difference, which becomes barely noticeable for $\mu_0 H = 4 \text{ T}$. The ferromagnetic component developing gradually below 100 K corresponds to $\approx 0.01 \mu\text{B}/\text{f.u.}$ at $T = 5 \text{ K}$.

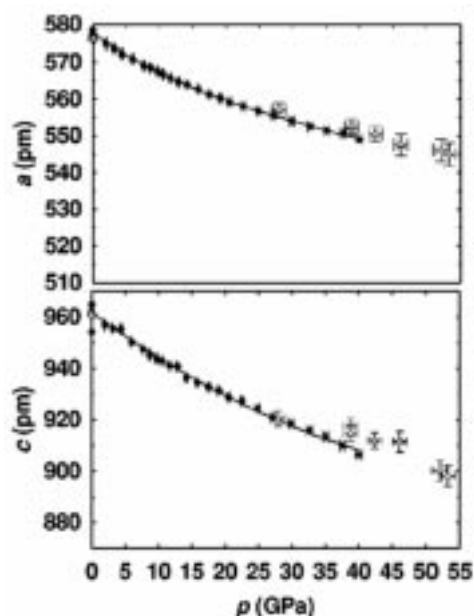
ters in random positions is appreciable for the high- T deposition, while low- T -deposited layers are magnetically “soft”. Finally, for the highest T_s (400°C), an anomaly around 50 K emerges, meaning probably a partial recovery of the bulk UN magnetism.

The second class relates to proposals that aim at complementary measurements on systems already extensively studied to shed light on some open questions. These experiments can be conducted on samples provided by the user or prepared at ITU for specific measurements. In this class of projects, there is a very high demand for high-pressure experiments (more than 20% of the demand and 15% of projects granted).

5f delocalization of UPd₃ under pressure

UPd₃ is the only known U-based binary intermetallic with localized 5f states. As the 5f localization leads to a large atomic volume expansion, it can be affected by external pressure. Recent ab initio calculations predicted that the 5f delocalization should occur around $p = 25 \text{ GPa}$ [7]. From the experiment carried out within the Actinide User Laboratory programme, it was demonstrated that no volume collapse nor anomaly occurs up to 53 GPa (see Fig. 5).

The 5f electronic states can be assumed to remain localised up to the pressure range achieved and therefore the qualitative predictions of the new SIC-LSD calculations have to be taken with caution.

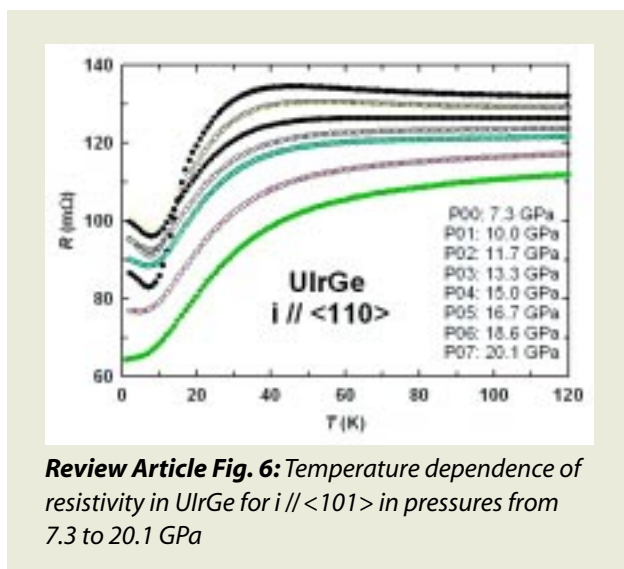


Review Article Fig. 5: Pressure dependence of the lattice parameters a and c . The full symbols were obtained from refinement of the data from the angle-dispersive mode, empty symbols from the energy dispersive experiment. The lines are quadratic fits through the data.

1. Basic Actinide Research

Suppression of magnetism and quantum critical phenomena in UIrGe under high pressure

A series of experiments was conducted on the well-known AnTX compounds displaying complex magnetic behaviours and addressing the interplay between magnetism and f -electron hybridisation. Indeed, to investigate the development of magnetism and possible quantum critical behaviour around 5 GPa in UIrGe, measurements of $R(T)$ dependence in various pressures up to 20 GPa were performed. In this system, the overall evolution of resistivity behaviour with pressure (see Fig. 6) can be attributed to gradual broadening of the $5f$ band.



Review Article Fig. 6: Temperature dependence of resistivity in UIrGe for $i // \langle 110 \rangle$ in pressures from 7.3 to 20.1 GPa

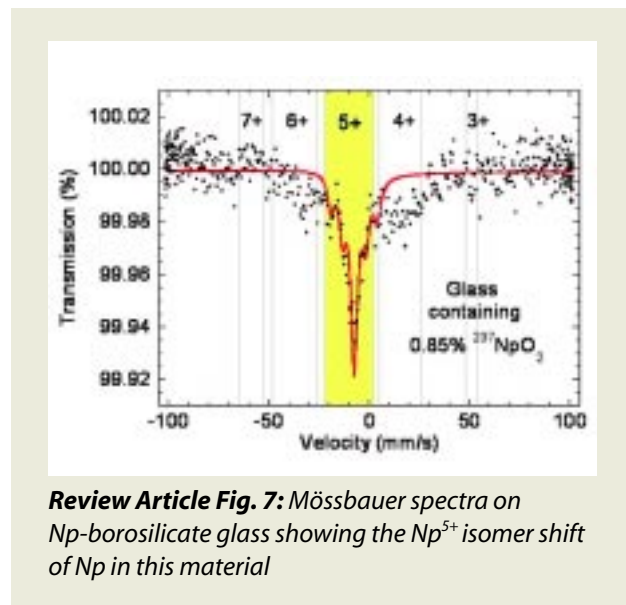
Within the same type of projects, the collapses of magnetism in UPtAl under pressure were studied, showing the dual role played by the $5f$ -ligand hybridisation and the investigations of UCoAl compound have shown that the most common explanation of the “Non Fermi Liquid (NFL)” mechanism as due to a close proximity to a quantum critical point is questionable. Indeed it was evidenced that in UCoAl the NFL character is a generic property related to the metamagnetic phase.

The final class summarises more applied-oriented problems.

Np environment in a borosilicate glass

This project addressed the question of the Np valence state in nuclear waste glasses. Our Actinide User Laboratory programme allows applied projects that do not dispose of all suitable characterisation tools (highly specialised such as Mössbauer) to have access to such facilities.

In this project, the valence state of Np could be unambiguously assigned to a $+V$ state (see Fig. 7).



Review Article Fig. 7: Mössbauer spectra on Np-borosilicate glass showing the Np^{5+} isomer shift of Np in this material

This information is important for further determination of the long-term behaviour (e.g. leaching properties) of such wastes.

Effect of alpha-emitter incorporation on the critical current density of high-temperature superconductors exposed to a magnetic field

This Danish project addressed the question of improving the performances of high- T_c superconductors in magnetic fields i.e. Bi-based superconducting cuprate ((Bi,Pb)₂Sr₂Ca₂CuO₁₀ (later called Bi2223)). One possible approach is to introduce columnar defects of diameter close to the coherence length of the superconducting electron pairs, i.e. in the nm range (these defects act as strong pinning centres for magnetic flux lines and prevent energy dissipation by thermally activated flux motion). This approach was already applied by means of heavy ions (Au) irradiation on Ag-sheathed Bi2223 composite tapes. Unfortunately, a large fraction of the ion beam energy is lost in the sheath and the creation of a high enough density of columnar defects necessitates a significant reduction of the Ag sheath thickness to values impracticable for the large-scale manufacture. Moreover, irradiation through the sheath material induces the activation of Ag nuclei, resulting in a very high and slowly decreasing radioactivity level. This disadvantage may be avoided by incorporating spontaneously decaying elements into the ceramic core. The userlab project pro-

posed a new original approach by incorporating (doping) an alpha-particle emitter (Am was selected) into a Bi2223. The effect of alpha particle irradiation on the magneto-transport properties of high-Tc superconductors is not known but these particles were shown to have penetration depth limited to a few micrometers in the superconducting matrix, which means that the radioactivity could well be confined within the sheath of the tape. The project started and preliminary experiments were conducted. In the first stage synthesis of doped material was addressed. Doped compounds were obtained but the pure phase could not be isolated. This project will certainly continue and preparation conditions should be optimised.

Impact of the Actinide User Laboratory

The first impact of the operation of the Actinide User Laboratory within the frame of the EC-Transnational Access to Research Infrastructures on our infrastructure has been to demonstrate that, despite restricted entrance requirements imposed by regulatory bodies due to security concerns, a User programme can be applied to a nuclear research institute. This successful experience has been used as a model to build up other pooled facilities i.e. the ACTINET-6 Network of Excellence.

This programme addresses directly the waning competence in nuclear field in Europe, a subject of considerable public concern. It has been a **major asset in the integration of new Member States research organisations** into ITU programmes. The use of the facility by scientists from the Candidate Countries was high – about 30% come from new Member States. Today, the Actinide User laboratory is attracting new collaborations, giving our research programme a **high visibility** and positioning the ITU as the **centre of excellence** in actinide research. Through this programme we have been involved in research areas which are not addressed institutionally, contributing to an increase of our global expertise and visibility of our own programmes. Moreover, access to the special facilities of the User Laboratory has enabled 30 different European team researchers (universities and national research centres) and their students to carry out research in the strategically important scientific field of basic actinide research to which they would have NO access without such opportunity, largely contributing to the **European Research Area** in the field. The eight Ph-D students that used the facility to achieve part of their experimental results, is a direct evidence of the capability of this programme to generate new original research.

Without the support provided by the EC-Transnational Access to Research Infrastructures Programme, this project would not have been possible. However, the long-term visibility of a user programme depends on obtaining funds for its continuation. We, therefore, are participating to EC-Access programme calls but, to ensure a continuous operation of our user facility, the JRC is also providing part of the funding from its Institutional budget.

Users potentially interested to participate to the programme should contact us at actuslab@itu.fzk.de or visit our website <http://itu.jrc.cec.eu.int>.

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1. Basic Actinide Research

To gain a complete understanding of fuel cycle activities, a thorough knowledge of the basic physical, chemical and material properties of actinides is essential. This knowledge is acquired by carrying out fundamental research projects that also gives our work an academic recognition. One of our major concerns is exploring the complexity of the behaviour of the $5f$ electrons in actinides, a task that requires unique experimental capabilities and challenges our understanding of matter.

Further tasks involve solution chemistry of the actinides, and two efforts centred mainly around the thermodynamics of the actinides, to critically evaluate the basic thermodynamic properties of the actinides and the measurement of high-temperature properties.

1.1 Solid State Physics of Actinide Metals and Compounds

Structural studies under pressure

Continuing our studies into the structural behaviour of the actinide metals with pressure using diamond-anvil cells and synchrotron radiation [1-4], we have completed new experiments on curium metal. Curium has been found to exhibit 5 independent crystallographic phases under pressure.

These studies performed in collaboration with Oak Ridge National Laboratory (USA) and the European Synchrotron Radiation Facility (ESRF, France) have shown that the initial double hexagonal close packed structure Cm I (space group, $P63/mmc$) transforms at 17(1) GPa to a face centred cubic Cm II (space group, $Fm3m$) phase. At a pressure of 37(2) GPa, curium then transforms to a monoclinic structure Cm III (space group, $C2/c$) which is a new structure previously unobserved throughout the actinide series of elements. At 56(4) GPa this latter phase converts to a face centred orthorhombic structure Cm IV (space group, $Fddd$) which, upon further pressure increase, undergoes a further transformation to form Cm V, a primitive orthorhombic structure (space group, $Pnma$) at 95(5) GPa. As in the case of its near neighbour americium, we observe that the $5f$ electron delocalisation in curium occurs in two steps although at considerably higher pressures, which was expected due to its $[Rn] 5f^7 6d^1 7s^2$ ground state electronic configuration. These high-pressure studies of curium were performed at room temperature using many different diamond-anvil

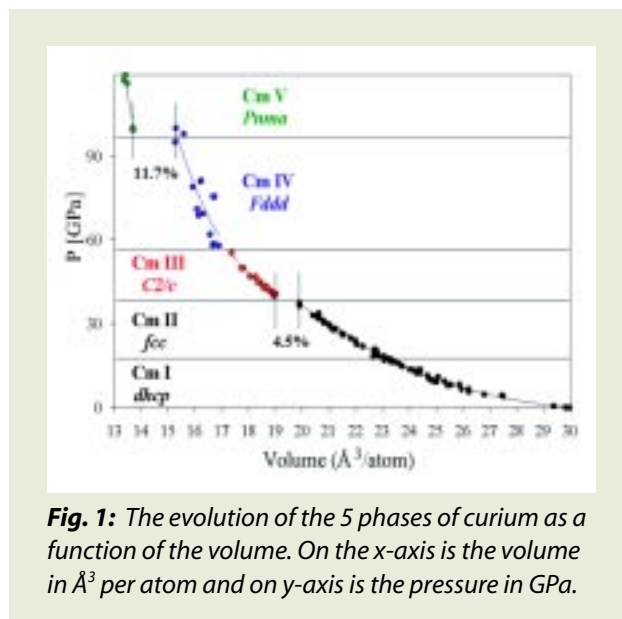


Fig. 1: The evolution of the 5 phases of curium as a function of the volume. On the x-axis is the volume in \AA^3 per atom and on y-axis is the pressure in GPa.

pressure cells [5]. The experiments were performed in an angular dispersive mode using synchrotron radiation of selected monochromatic wavelength at the ESRF. Figure 1 shows the pressure behaviour of curium, where the volume per atom at ambient pressure is plotted against pressure.

The most interesting new aspect of these studies is the formation of the Cm III phase starting at 37(2) GPa, which remains the stable structure until 56(4) GPa. Rietveld refinements have identified this phase as having a monoclinic structure with the space group $C2/c$, Cm on the $4e$ sites $(0, y, 1/4)$. Figure 2 shows the Rietveld refinement obtained for Cm III and a model of the structure. From the model it can be seen that in comparison to the Am III $Fddd$ structure, Cm III is composed of slightly distorted (rectangularly distorted) close-packed hexagonal planes, but in contrast to Am III, with a stacking which reduces the symmetry to monoclinic. At this pressure each Cm atom has 12 neighbours with atomic distances between 2.83 and 3.29 \AA .

A small collapse of about 4.5% in relative volume at the onset of the Cm III phase can be seen in Fig. 1 indicating a partial $5f$ electron delocalisation.

The Cm V phase observed above 95(5) GPa is isostructural with the recently discovered Am IV phase [1] and Rietveld refinements establish this structure as primitive orthorhombic (space group $Pnma$, Cm on the $4c$ sites $(x, 1/4, z)$). This transition from Cm IV to Cm V is also accompanied by a larger volume collapse of about 11.7%.

1. Basic Actinide Research

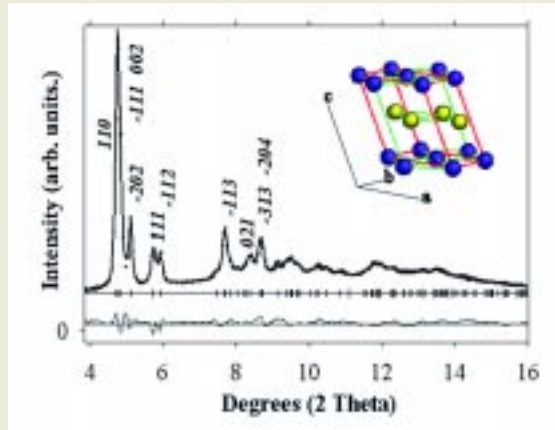


Fig. 2: Rietveld refinement of Curium III at 45 GPa showing calculated reflection tick marks, principal reflections and difference profile. The insert shows a model of this monoclinic (s.g. C/2c) structure.

Taken together the 2 collapses mentioned above represent the appearance of itinerant 5f electrons in the metallic bonding with the delocalisation occurring in 2 stages as was previously observed with americium.

The mysteries of low-temperature ground states of actinide oxides resolved by synchrotron experiments

The discovery in 2002 [6] of quadrupole ordering in the NpO_2 opened a new chapter in our effort to understand the low-temperature ground states of actinide oxides. We concentrate in this report on the solid solutions $(\text{U}_{1-x}\text{Np}_x)\text{O}_2$, all with the simple cubic CaF_2 crystal structure and which have been studied for many years. The phenomenon found at 25 K in NpO_2 ($x = 1$) is that the 5f charge distribution develops an anisotropic component that exhibits *long-range ordering* at $T_0 = 25$ K. This is known as the ordering of the charge *quadrupoles*.

The experiments consist of tuning the photon energy to the actinide M absorption edge (at which photon energy core 3d electrons are promoted to the partially occupied 5f valence states), and then, once the repeat of the anisotropic charge distribution is known, to measure the azimuthal distribution of the scattered resonant x-ray intensity. This intensity distribution is related to the symmetry of the 5f anisotropic charge distribution. The experiments were performed at the European Synchrotron Radiation Facility (ESRF) in Grenoble, France, with the single crystals mounted in ITU.

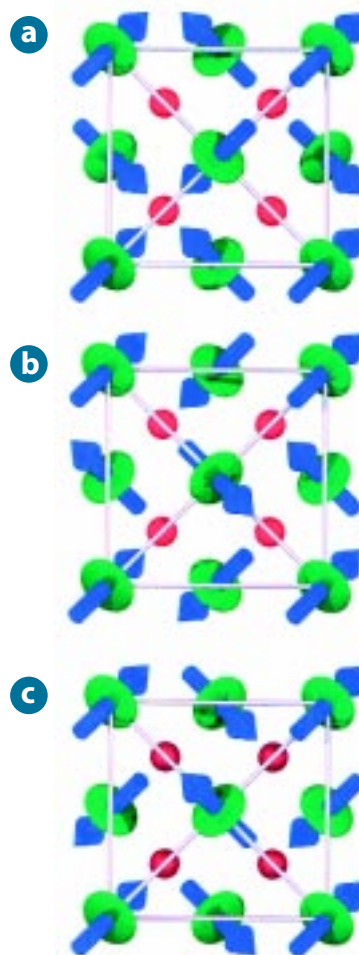


Fig. 3: Schematic representation of triple-k magnetic and electric-quadrupole ordering within the a - b plane for the single longitudinal (a) and two transverse (b,c) modes. The magnetic (dipole) moments are represented by blue arrows whereas the electric-quadrupole moments are shown as the green ellipsoids. The red spheres represent oxygen atoms.

NpO_2 has pure quadrupole ordering, with no measurable dipole component. In 2003 experiments were performed [7] on a single crystal with $x = 0.25$. Dipole ordering was found on both the U and Np ions, and quadrupolar ordering was also found associated with both ions. However, these were different from those in pure NpO_2 . Whereas a longitudinal (L) configuration was found for $x = 1$ (NpO_2), the $x = 0.25$ sample was found to have a transverse (T) configuration. The difference between these configurations is shown in Fig. 3.

The understanding of these different configurations then allowed an experiment to be performed in 2004 on a single crystal of UO_2 ($x = 0$). The azimuthal dependence of the intensity from the (112) reflection in UO_2 is shown in Fig. 4, together with the predictions for a T configuration. The complication in UO_2 is that both dipole ordering *and* an internal distortion of the oxygen atoms due to the quadrupole ordering are present; these have contrived to make it difficult to observe directly the quadrupole ordering in UO_2 , although such ordering was predicted almost 40 years ago [8], and the oxygen displacements reported 30 years ago [9].

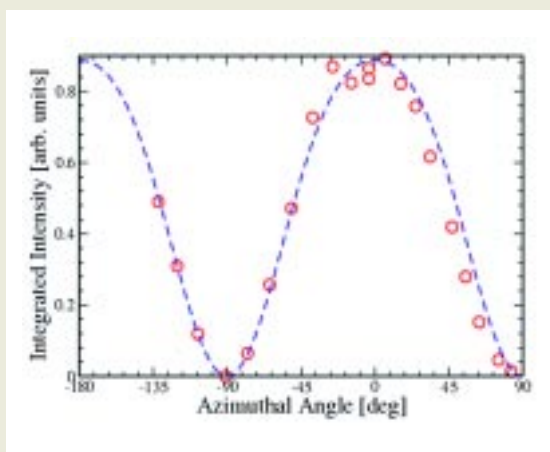


Fig. 4: Integrated intensity as a function of azimuthal angle for the (112) superstructure reflection in the sigma-sigma polarisation channel (open circles). The broken line is a fit to the model for quadrupolar ordering (see text for details). In the model there is an incoherent superposition of the two T modes.

These experiments also suggest a qualitative explanation of the unusual behaviour [10] of the mixed oxides with $0.40 < x < 0.80$. In this region there is competition between the L and T quadrupole ordering, resulting in quadrupolar *frustration* and only *short-range* dipole and quadrupole ordering. Experiments to detect such effects are planned.

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1.2 Electronic Structure of the Actinide Hydrides (AnH_3): A Photoemission Study

The evolution of the electronic structure within the actinide series shows the general trend for $5f$ localisation with increasing atomic number and a transition takes place between Pu and Am. However, even for “simple” metals, there are still unsolved questions such as the absence of magnetism in δ -Pu. Further insight into the electronic structure can be obtained by extending the study to series of simple compounds (nitrides, sulphides, etc), where the electronic structure is further modified by a supplementary ligand element. We report such a study performed on the actinide hydride series (AnH_3 with $\text{An} = \text{U}, \text{Np}, \text{Pu}$ and Am).

The actinide hydrides are highly reactive. Therefore they were prepared in situ by reactive sputter deposition of the actinides in an Ar- H_2 atmosphere. The high concentration of atomic hydrogen in the plasma ensures the formation of the highest actinide hydrides, i.e. AnH_3 . Figure 5 compares the photoemission valence band spectra of actinide metals and their corresponding hydrides.

The spectra of U, Np and Pu metal are all similar, showing the major intensity, of f -character, at the Fermi-level. This directly proves the itinerant character of the f -states, which, together with the spd states, form a conduction band. In Am metal the f -states are withdrawn from the Fermi-level and show a multiplet final state structure, characteristic for localised states.

1. Basic Actinide Research

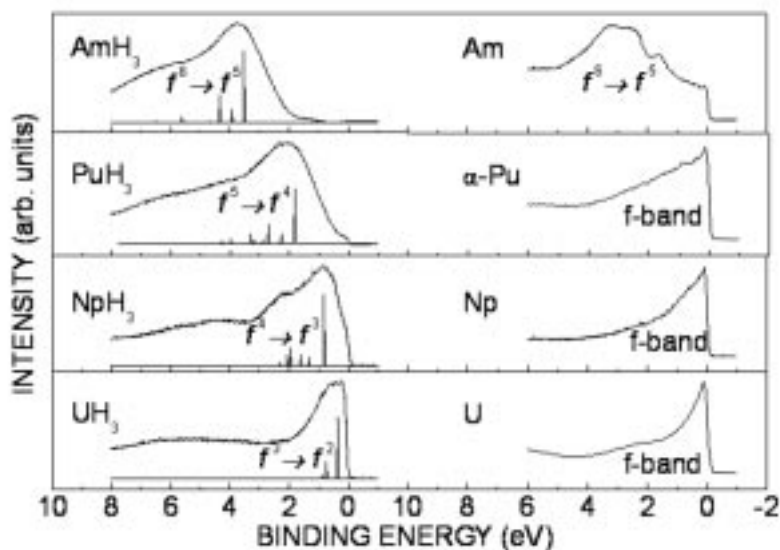


Fig. 5: Valence band spectra of actinide metals and their corresponding hydrides.

In the hydrides case, the f -states are shifted to higher binding energy, indicative of their localisation. In general, the presence of hydrogen, squeezed into interstitial positions, pushes the metal (d or f) states towards localisation because of the lattice expansion and the partial ionic character of the metal-hydrogen bond. The peak shapes are well described by multiplet transitions ($f^n \rightarrow f^{n-1}$), and the respective ground-state configurations (f^3 to f^6 from U to Am) are consistent with the magnetic bulk properties. In UH_3 [1], the multiplet is located right at the Fermi-level. This is a weakly localised configuration, whose ground-state may still be itinerant. UH_3 is an itinerant ferromagnet. PuH_3 has an effective moment of $0.76 \mu_B$, which is attributed to a $5f^5$ configuration. AmH_3 has no magnetic moment, consistent with the $5f^6$ ground-state configuration and cancellation of spin and orbital moment. Interestingly, AmH_3 has no emission at the Fermi-level, so it is an insulator. It displays characteristic rare-earth behaviour, for which the tri-hydrides are also insulators due to the transfer of all three conduction band states in the H-1s anion band.

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2. Spent Fuel Characterisation

During the reporting period, ITU has continued working along the guidelines defined for FP6 pursuing a trend towards tighter convergence and integration of the activities both in-house and in the frame of collaboration with external partners. The various sections of this chapter provide examples and highlight of experimental activities, all related to a spent fuel direct disposal scenario, which can schematically be grouped along four main lines of study.

- studies on alpha-radiolysis on one hand and on the hydrogen suppressing effect on the other, considered as opposite factors in determining the redox conditions ultimately governing the fuel corrosion process;
- characterization of the corrosion process of spent fuel: source term, burnup, irradiation history effects, corrosion mechanisms and dissolution rates.
- analysis of the role of plutonium in the overall corrosion process of spent fuel;
- investigation of alterations occurring in the spent fuel during storage prior to water access and their possible influence on the interaction with groundwater;

It is foreseen to extend the scope of these investigations to cover other concepts of wasteform, like e.g. Minor Actinide (MA) conditioning matrices, and irradiated fuel from advanced reactors/fuel cycles.

Most ITU activities are embedded in an international, converging effort, which sees the main European institutions in the field participating in common projects to define and assess input for performance assessment exercises for European repositories. This effort was represented by the EC Shared Cost Action *Spent Fuel Stability under Repository Conditions*, which ended in 2004. This project aimed at developing and validating a model to predict the radionuclides release rate from spent nuclear fuel which can thereafter be used in the performance assessment exercises for repository systems. Our partners in this project were 8 international institutions from 5 European countries. ITU contributed to this project with important studies on radiolysis, hydrogen effects, and instant release fraction studies, which were used for the development of the MAM code [1].

In 2004 the integrated project NF-PRO also started, which is aimed at understanding and modeling key processes and their coupling in the near field, for different host rocks and repository strategies. Results and conclusions will be integrated in performance assessment.

HIGHLIGHT

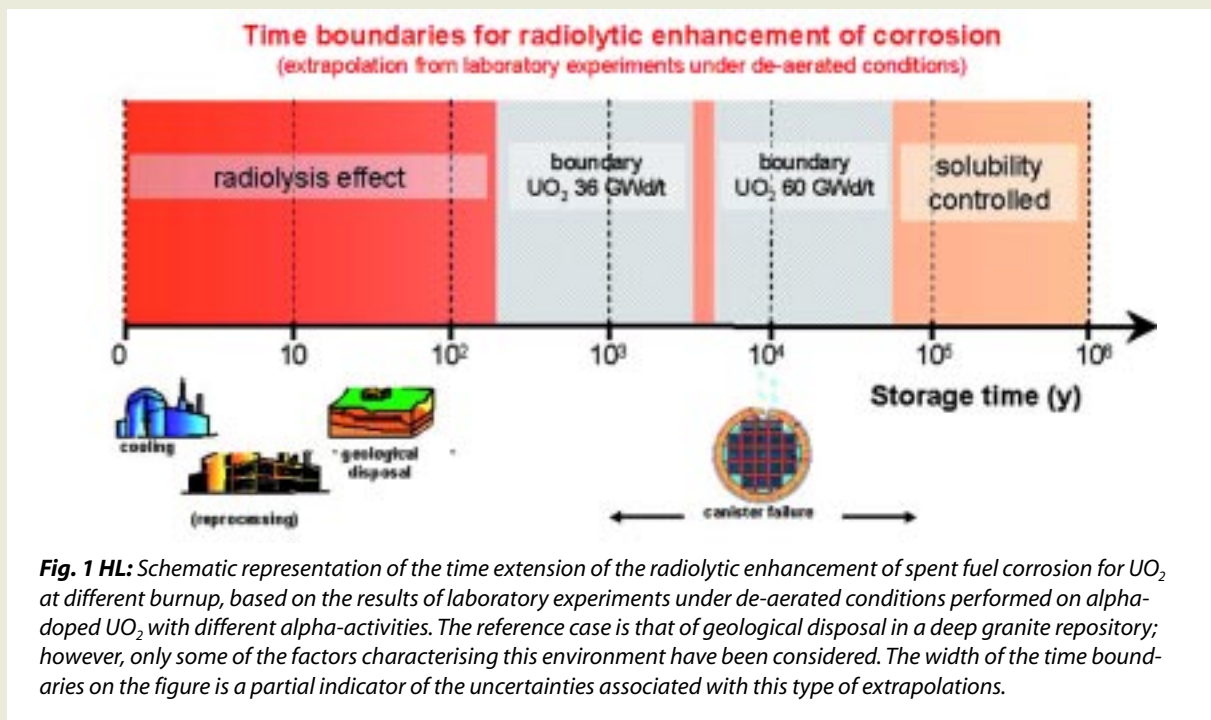
Limits of the Radiolytic Enhancement of Spent Fuel Dissolution in Groundwater: Activity Dependence

Alpha-radiolysis (the ionising effects of alpha particles) constitutes an important factor that may affect the corrosion behaviour of spent fuel exposed to groundwater in a geological repository. Such a repository is characterized in all European concepts by the absence of oxygen or oxidants, which could enhance fuel corrosion.

The dissolution of uranium oxide occurs via chemical dissolution of U(IV) in absence of oxygen, and via oxidation-dissolution of U(VI) under oxidising conditions. The oxidative dissolution mobilizes several orders of magnitude more uranium than chemical dissolution. As a result of the radiolysis of water, molecular and radical species are formed near the surface of the fuel. The oxidising species generated by radiolysis may oxidise the surface of the uranium dioxide fuel causing an increase of the overall corrosion rate of the fuel matrix. Alpha-radiolysis is the mechanism of interest, since in old spent fuel the alpha-emission will dominate the radiation field in and around the fuel.

During the past 6 years, an extensive experimental effort was conducted at ITU to study the amplitude and extent of the alpha-radiolysis effect on UO₂ matrix corrosion (see e.g. review article, TUAR 2003, pp.12-19 or [1-3]). UO₂ pellets containing alpha-emitters ²³⁸Pu or ²³³U (so-called alpha-doped UO₂) were fabricated in-house using sol-gel techniques to ensure a homogeneous distribution of the alpha-decay events in the matrix. By varying the fraction or the type of alpha-emitter, a spectrum of materials spanning over 5 orders of magnitude of specific alpha-activity was obtained. The highest activity (~10¹⁰ Bqg⁻¹) simulates irradiated MOX at discharge, while the lowest activity (~10⁶ Bqg⁻¹) would correspond to the same fuel after ~10⁵ y of storage (see Fig. 1 HL).

The first experiments, focused on the high activity materials, demonstrated the existence and effects of enhanced corrosion due to radiolysis. In recent years, the focus has shifted to the determination of the limits of this radiolytic effect, in terms of its dependence on



the fuel activity and of possible interaction with other environmental parameters acting in the repository system (see this chapter)¹.

In particular, an effort was devoted to determining the alpha-activity limit below which the radiolytic contribution to the overall corrosion behaviour is negligible compared to other environmental effects. The alpha-activity threshold below which fuel dissolution is governed by the redox condition of the leaching environment, hence is negligible under anoxic conditions was determined under a defined range of experimental conditions. Leaching tests at room temperature in deionized and granite groundwater under anoxic and deaerated conditions showed that there is essentially no difference between the leaching behaviour of undoped depleted UO_2 and that of UO_2 containing 1wt% of ^{233}U , whereas a clear radiolytic enhancement of fuel dissolution was observed in the case of alpha-doped UO_2 containing 10 wt% of ^{233}U . In terms of the age of simulated spent fuel, as graphically illustrated in Figure 1, this would correspond to saying that no radiolytic enhancement of dissolution would be expected for spent fuel with an age, $t > t_{threshold}$. For high burnup UO_2 or for MOX fuel, approximately $10^4 y < t_{threshold} < 10^5 y$ of storage. In the case of low-burnup UO_2 fuel, $t_{threshold} < 10^4 y$.

However, the experimental systems considered to determine this activity threshold represent only partly the whole spectrum of conditions characterizing the spent fuel repository. More work is ongoing to fully characterise the effects of radiolysis, together with other relevant processes acting in the geologic repository.

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¹ These aspects, relevant as input for performance assessment exercises for European repositories, have partly been pursued in the frame of ITU's contribution to the EC-funded project Spent Fuel Stability (Contract No. FIKW-CT-2001-20192 SFS) and in the frame of bilateral collaboration between ITU and ENRESA-CIEMAT of Spain.

2. Spent Fuel Characterisation

40 partners from 10 European countries participate to this new project. ITU contributes with activities concerning the grain boundary inventory of spent MOX fuel as well as the accumulation of α -decay damage in doped UO_2 and unirradiated MOX. The corrosion behaviour of spent fuel in defect rodlets in water vapour (90°C) under $\text{Ar} + \text{H}_2$ is studied in collaboration with SKB. Leaching experiments on spent fuel with different burn-up are carried out in collaboration with ENRESA.

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2.1 Redox Conditions, Hydrogen Effect and Radiolysis

H_2 overpressure suppressing effect on fuel corrosion

The experimental campaigns aimed at studying the H_2 overpressure effect on fuel corrosion (collaboration between ITU and SKB, Sweden), simulating conditions expected in a deep geological repository due to water corrosion of iron canisters. These studies are performed

using airtight autoclaves placed in a glove box or hot cell [1]. The current effort is focused on testing irradiated fuel: an experiment with 47.5 GWd/t MOX, started in 2002, is still running at the nominal maximum H_2 pressure of ~52 bar. The new results confirm the suppression of oxidative fuel matrix corrosion (TUAR 2003, pp.12-19), considering trends for relevant fission products and actinides. Particularly interesting is the measured evolution with time of the Fraction of Inventory in the Aqueous Phase (FIAP) of cesium. This quantity leveled off after ~1.5 y of leaching at low values never observed before. This seems to provide a striking indication that under 50 bar of H_2 the corrosion of irradiated fuel has essentially stopped.

Previous experiments were performed on alpha-doped UO_2 . A 10wt-% ^{233}U -doped UO_2 pellet simulating the radioactivity of real spent fuel that is ~ 10^5 year old was used.

An average dose rate of 83 Gy/h was dissipated in the 10^{-2} M NaCl + $2 \cdot 10^{-3}$ M HCO_3^- aqueous leachant. This experiment lasted 2.2 years. $[\text{H}_2]$, $[\text{O}_2]$, E_h and temperature were measured at given time intervals, while the leachate was regularly analyzed by different techniques to determine the concentration of uranium.

The results are summarized in Fig. 1. It can be concluded that no oxidative corrosion occurred of the stoichiometric ^{233}U -doped UO_2 pellet in contact with dissolved H_2 in the concentration range of 10^{-2} – 10^{-5} M. An eventual threshold for this corrosion-inhibiting effect would have to be $<10^{-5}$ M $[\text{H}_2]$. The average concentration of U in the

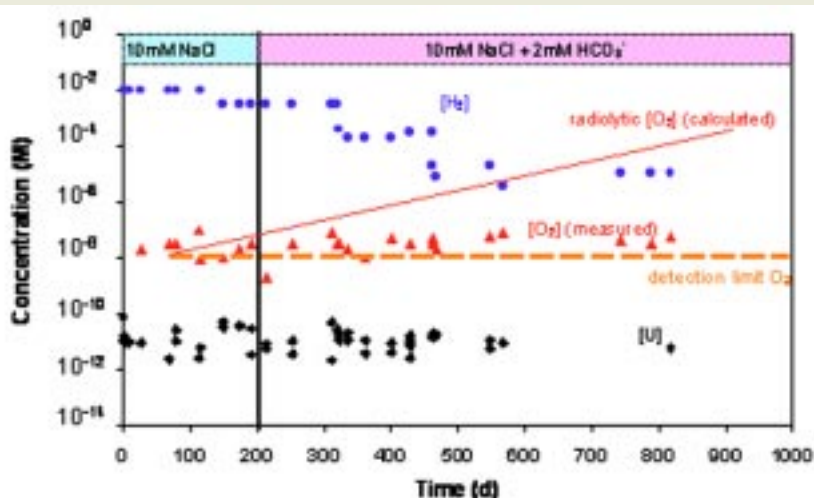


Fig. 1: Concentrations of U, H_2 and O_2 dissolved in the leachate as function of time. The calculated concentration of O_2 produced by radiolysis is also plotted (solid line). The dashed horizontal line represents the detection limit for dissolved O_2 determination; the vertical dotted line indicates the time at which 2mM carbonate was added to the leachant.

leachate was $1.5 \cdot 10^{-11}$ M. The O_2 concentration in the leachant remained essentially constant, in spite of the continuous radiolytic production of oxygen. The fate of the “missing” oxygen is not clear. A post-test characterization of the fuel pellet showed no signs of corrosion on the fuel surface. Deposits of titanium oxide were found in the vessel.

The knowledge gained so far from these experiments converges to the conclusion that the hydrogen effect will have a significant impact on the source term release and therefore a strong influence on the performance assessment of the nuclear fuel repository.

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Leaching of alpha-doped UO_2 in presence of hydrogen

Static batch leaching experiments in granite groundwater were performed on discs of 10% ^{233}U -doped UO_2 and of undoped UO_2 . The total duration of the test was 1 year.

The pH and E_h were monitored during the experiments. A constant flow of $Ar/(6\%)H_2$ bubbled in the water during the experiments. After sealing the reactor vessel, the fuel sample remained suspended in a quartz sample holder above the water. The sample holder was immersed in the leachant when the water had reached sufficiently low Eh values ($-400 \div -500$ mV_{SHE}). Aliquots of leachate were extracted at given time intervals and analyzed by HR-ICP-MS.

Figure 2 shows the concentration of ^{238}U and ^{233}U in solution as a function of time. For both undoped UO_2 and alpha-doped UO_2 no further corrosion could be observed after the initial release. The initial concentration of ^{238}U in solution was higher for UO_2 with 10% ^{233}U than for undoped UO_2 . This can be attributed to the surface pre-oxidation conditions developed before sample immersion. However, no increment of the dissolved amounts occurred after the initial release. The measured concentrations in solution actually tended to *decrease* after longer leaching times. The analysis of the $^{233}U/^{238}U$ ratio in solution indicated that congruent dissolution of the material occurred. The E_h potential measured in the solutions showed a general increasing trend with time, but remained negative throughout the duration of the experiment [1]. The observed trends indicate an essential suppression of the radiolytic enhancement of corrosion, which was observed for the same alpha-doped material under anoxic/unaerated conditions in absence of hydro-

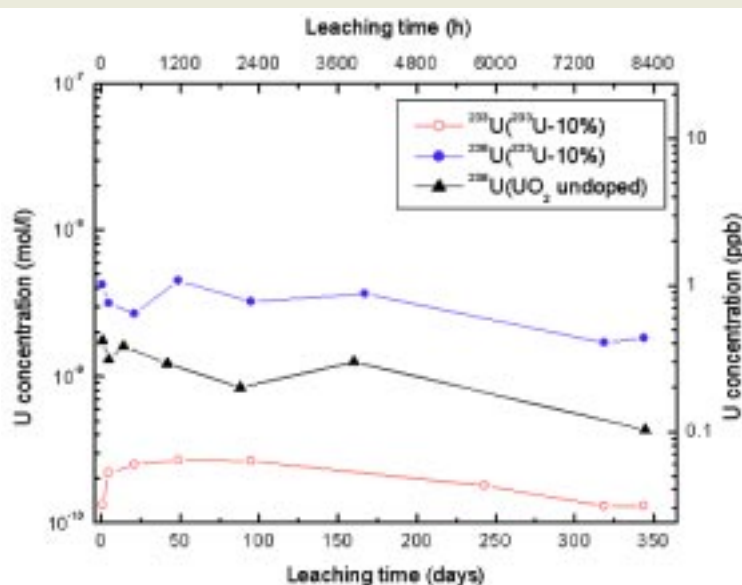


Fig. 2: Concentration of U vs. leaching time in experiments performed under $Ar/(6\%)H_2$ in granite groundwater ($pH = 7.5$)

2. Spent Fuel Characterisation

gen [2]. This work has partly been performed in collaboration with ENRESA and CIEMAT, Spain.

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Electrochemical testing of alpha-doped UO_2

Electrochemical long-term testing under static conditions was carried out on vacuum-impregnated and polished 1% ^{233}U -doped UO_2 at room temperature. The S/V ratio was in the range of 0.03 - 0.07 m^{-1} . The experiments were carried out in an electrochemical cell under inert gas purging. The gas N_2 or N_2/H_2 was purified using an oxygen trap.

The free corrosion potential of the UO_2 and the E_h of the 10 mM NaCl solution were followed using commercially available reference and combination redox electrodes. The solution (100 ml 10^{-3} M NaCl) was bubbled for 24 h to minimize the oxygen content. Later only the gas phase

above the electrolyte was purged to avoid potential fluctuations generated by gas bubbles. After some 100 hours the purging gas was switched from N_2 to $\text{N}_2/(8\% \text{H}_2)$ to study the influence of H_2 .

Results of the potential measurements are shown in Fig. 3. The measured E_h dropped by about 450 mV after switching from purging with N_2 to $\text{N}_2/(8\%)\text{H}_2$ indicating that the redox conditions had become more reducing. After a delay, possibly caused by the consumption of remaining oxygen at the UO_2 surface, switching to $\text{N}_2/\text{H}_2(8\%)$ led to a drop of 200 mV also in the corrosion potential that was thereafter very slowly decreasing. The corrosion potential drop represented a decrease of the corrosion rate of about 2 orders of magnitude.

Dynamic leaching of spent UO_2 fuel under oxidising conditions

The aim of this study, (collaboration with ENRESA and Polytechnic University of Catalunya, Spain), is the determination of dissolution rates of spent fuel in the absence of secondary phase precipitation.

A flow-rate of about 0.06 g/min constitutes the minimum that can be adopted with our continuous flow-through dynamic leaching setup without losing reproducibility of the results. However, the first tests on UO_2 with 53 MWd/kgU samples revealed that even at this low flow-rate the concentration of many nuclides was below

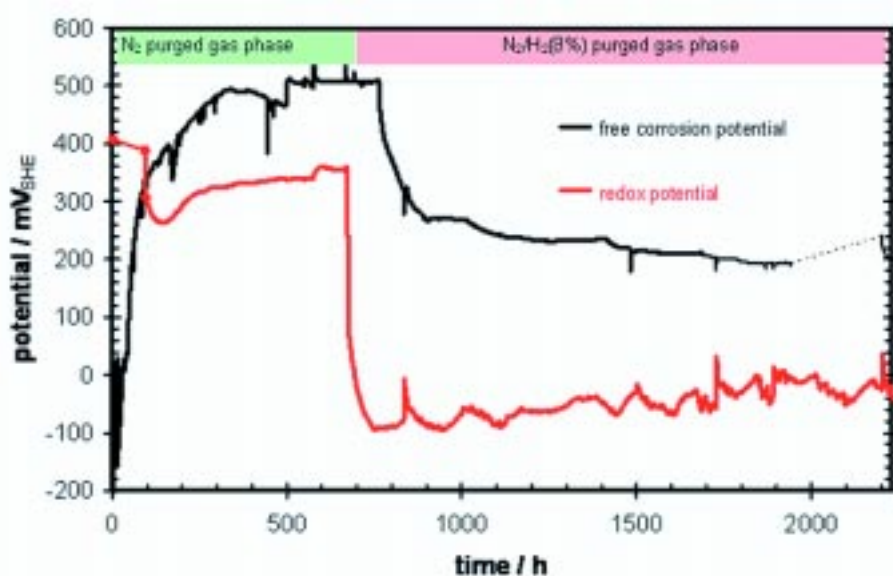


Fig. 3: Long-term corrosion test of 1% ^{233}U -doped UO_2 in 0.01 M NaCl-solution. Free corrosion potential and redox potential as a function of time. The gas phase in the vessel was purged with N_2 for the first 672 h, then the purging switched to $\text{N}_2/8\%\text{H}_2$.

the detection limit of the high resolution ICP-MS and gamma-spectrometry used to analyse the leachate solutions. A ~100-fold increase of the surface area of the samples solved this problem. Thus, a new set of experiments using milled pellets of the same fuel was carried out.

The leachant used was water containing 10^{-3} M NaHCO_3 and $1.9 \cdot 10^{-2}$ M NaCl . The tests were performed in air, so atmospheric oxygen established oxidising redox conditions. The experiment duration was ~5 months. The results are shown in Fig. 4 as ΔFIAP vs. leaching time [1]. The fuel inventory was obtained using the KORIGEN code. Initially, relatively high flow-rates were set to fill the reactor. The nominal flow-rate for the leaching experiment was 0.06 ± 0.01 g/min.

Congruent dissolution of Np with U was observed. Lower dissolution rates were obtained for Pu (14 times smaller than U) while higher rates were found for Sr and Cs (2 and 5 times higher than U, respectively). These results are in line with literature data [2].

The valley shown in Fig. 4 between approximately 60 and 115 days represents the effect of an accidental stop of the leachant flow. The recovery of the previous ΔFIAP levels after returning to the nominal flow-rate gives indication that steady-state conditions have been reached,

thus confirming the suitability of the set-up and the reliability of the results under nominal conditions.

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2.2 Studies on the Corrosion Behaviour of Pu-containing Fuel

Dissolution and precipitation in presence of radiolysis products

H_2O_2 is a radiolysis product that reacts with UO_2 and leads to the precipitation of secondary studtite-type minerals $\text{UO}_4 \cdot 2 \text{H}_2\text{O}$ and $\text{UO}_4 \cdot 4 \text{H}_2\text{O}$ [1]. Peroxo compounds $[\text{Pu}_2(\text{O}_2)_3]^{2+} \cdot \text{Y}^{2-}$ were synthesized in strongly acidic solutions [2]. Experiments in our laboratory inves-

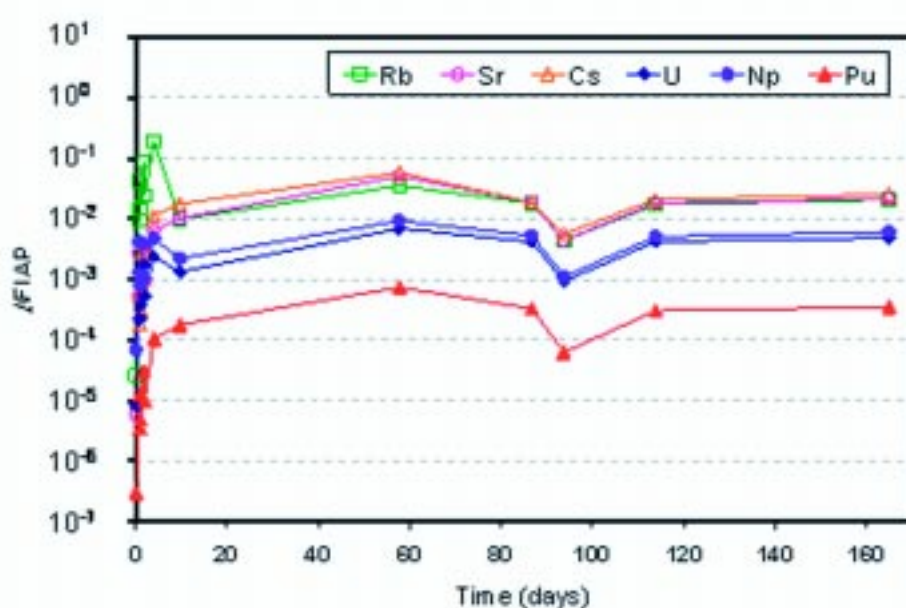


Fig. 4: Dynamic leaching of spent fuel (UO_2 , 53 MWd/kgU). Fraction of Inventory in the Aqueous Phase vs. time

2. Spent Fuel Characterisation

tigated if peroxy compounds of Pu could be formed under neutral or slightly acidic conditions, and if these phases could be incorporated into studtite-type phases of uranium.

UO₂ and PuO₂ were immersed in aqueous solutions containing H₂O₂. After ~6 days, the formation of UO₄ peroxy phases was observed and confirmed by Raman spectroscopy. PuO₂ samples did not show any secondary phase formation or surface alteration. It proved that the Pu peroxy compound cannot be obtained in neutral or slightly acidic media. Differences of the crystal structures of both peroxides, and the necessity for inclusion of counter-anions in the case of the Pu compounds, lead to the conclusion that the combined formation of both secondary products during storage is unlikely.

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Corrosion of unirradiated MOX fuel in groundwater

The surface of leached unirradiated MOX, UO₂ and PuO₂ pellets was characterized by Scanning Electron Microscopy (SEM). The MOX materials were fabricated via sol-gel to obtain a homogeneous distribution of plutonium within the UO₂ matrix. Sequential leaching experiments were performed at room temperature in granite groundwater, under unaerated condition. The S/V ratio, calculated using the geometric surface, was ~2.5 m⁻¹. The leaching duration was approximately 1 year. Figure 5 shows SEM micrographs of the leached surfaces after 1 and 8000 hours of leaching. No significant etching/precipitation is evident on any leached surface of MOX and of PuO₂, whereas etching and schoepite reprecipitation are evident in the case of UO₂ [1,2].

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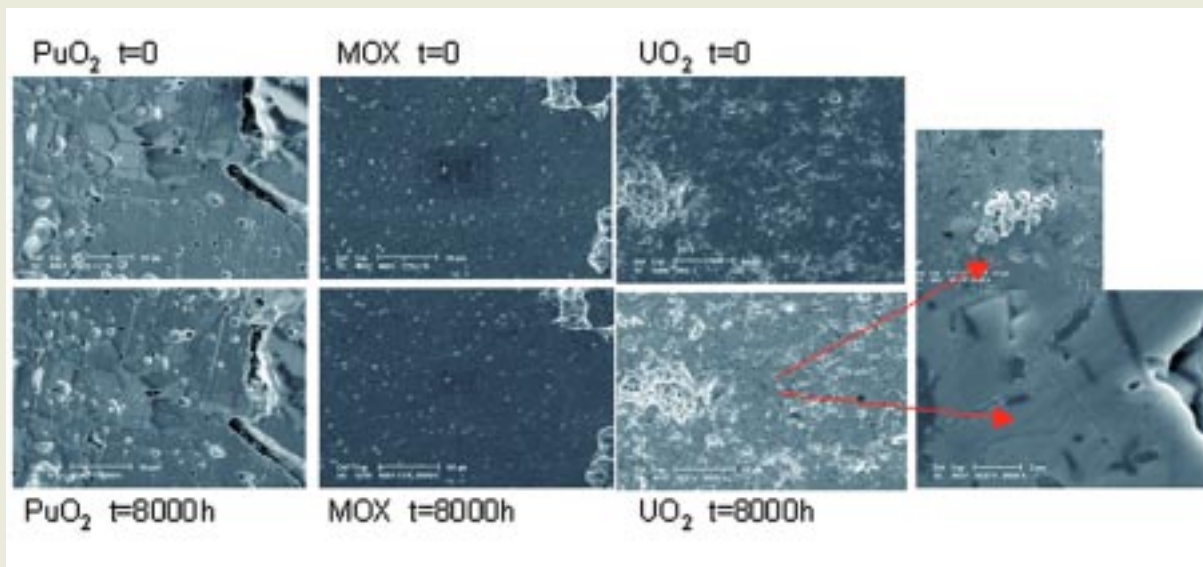


Fig. 5: SEM micrographs after 1 and 8000 hours of leaching in granite groundwater for PuO₂, MOX containing ~10 wt. % ²³⁹Pu, and UO₂. No significant alterations of PuO₂ and MOX surfaces were visible, while reprecipitation and grain boundary etching (indicated by the red arrows on the figure) occurred during leaching of UO₂.

2.3 Effect of Alpha-decay on Long-term Behaviour of Irradiated Fuel

Corrosion of spent nuclear fuel can be regarded not only in terms of chemistry, but also from a mechanical point of view. The long term accumulation of decay damage in the fuel structure, possibly enhanced by precipitation at grain boundaries of helium [1] may affect the structural integrity of the fuel. The point defects created by collision cascades precipitate in extended defects like dislocation loops and voids that are source of internal stresses within the polycrystalline structure. The extreme end-effect could be a grain separation process (TUAR 2003, p.12-19; TUAR 2001, p. 69-71). Furthermore, this type of fuel alteration could negatively affect an eventual handling of the spent fuel after interim storage and might ultimately influence the dissolution process, e.g. by increasing the surface of the fuel exposed to water after failure of the confinement barriers. Together with the worst-case scenario associated with decay damage and stress accumulation in high alpha-activity materials, there is also evidence indicating that in other cases no loss of mechanical integrity occurs at elevated accumulated decay doses. This is the case e.g. of the SUPERFACT fuel, a special irradiation of UO_2 containing MA which

ended in 1991. Ceramographic and SEM examination of UO_2 containing 20% Am and 20% Np after 13 years of storage (with an accumulated dose corresponding to 50 GWD/t MOX fuel after 10^6 years of storage) revealed no visible changes in the fuel structure compared to the morphology observed in 1991.

The structural modifications occurring during very long storage times must be anticipated from the results of calculations and laboratory experiments. Some additional information may be provided by natural analogues. However, no direct evidence of the actual behaviour of spent fuel during extended interim or long term storage is available.

Sputtering and ballistic ejection of atoms in water

An aspect correlated to the specific surface area of the fuel and linked to the alpha-decay process is the direct sputtering of the radioactive material into water contacting the fuel. In this case the radionuclide mobilization is determined only by ballistic mechanisms.

Figure 6 shows a normalized distribution of the recoils and of the sputtered matrix atoms calculated using SRIM2000 [2]. An input file with an anisotropic distribution of alpha-recoils of 101 keV was used for the calculation. The quantity of U or Pu that could be sputtered

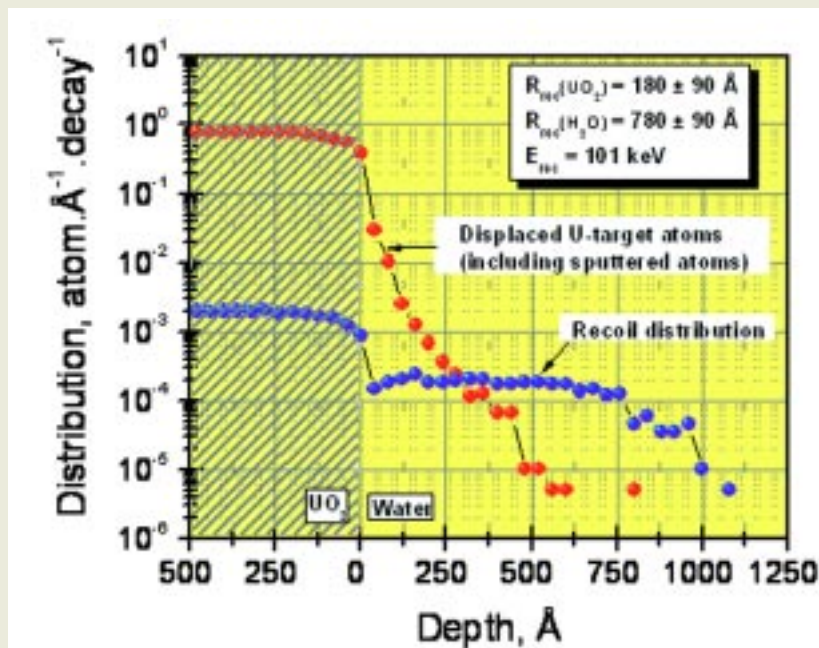


Fig. 6: Distribution of recoil atoms and sputtered atoms from a UO_2 layer calculated with the SRIM2000 code [2]. The distribution refers to a surface layer of 500 Å and assumes that thickness and surface of the solid exposed to water are $\gg 500$ Å.

2. Spent Fuel Characterisation

from an infinite surface was assessed. Only a layer of thickness corresponding to the range of a typical recoil nucleus contributed to the release. For an activity of $3.8 \cdot 10^{10}$ Bq/g, corresponding to irradiated MOX at discharge, the amount of sputtered atoms is around $1 \mu\text{g}/\text{m}^2\text{d}$, approximately 3 orders of magnitude less than the dissolution rate measured for the same material in leaching experiments [1]. The sputtered amount would be proportionally smaller for lower activity, i.e. for older spent fuel. It can therefore be stated that the direct sputtering plays a negligible role in the release of radionuclides in case of fuel exposure to water if the fuel retains its structural integrity. In case the fuel would turn into powder this picture might change drastically since the surface over volume ratio would increase allowing a larger fraction of the recoil atoms to be close enough to surfaces to be directly sputtered into the water.

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3.1 Commissioning Tests on a Shielded Secondary Ion Mass Spectrometer for the Analysis of Irradiated Nuclear Fuel

The new CAMECA IMS 6F secondary ion mass spectrometer (SIMS) for the analysis of irradiated nuclear fuel has a number of special features for the protection of the laboratory personnel. These include a stainless steel glove box to restrict the spread of radioactive contamination during sample transfer and a lead cell with walls 10 cm thick surrounding the glove box. The lead cell limits the radiation dose rate received by the operator to $5 \mu\text{Sv}\cdot\text{h}^{-1}$ when handling irradiated nuclear fuel samples of activity up to 75 GBq. Nuclear fuel samples will be transported to the machine in a La Calhène container inside a lead transport flask and introduced into the steel glove box through a port in the side of the lead cell. The introduction of the sample in the analysis chamber is remotely controlled with possibility of manual operation in case of malfunction. Another key feature of the SIMS is the immersion lens, the contamination level of which is continuously monitored by real time integration of the dose equivalent to the amount of material sputtered during analysis. The immersion lens is modified to facilitate its removal for cleaning when the contamination level is equivalent to a dose of $2 \text{mSv}\cdot\text{h}^{-1}$.

Commissioning tests have been carried out on unirradiated material. The objectives are: (1) to evaluate the performances of the shielded machine by comparison with similar measurements performed on a non-shielded IMS 6f; (2) to develop working procedures for the analysis of irradiated materials. The commissioning tests started with the detection of the hydride phase in Zircaloy cladding and showed that at very high mass resolution (18000) Zr-hydride molecular species can be discriminated from Zr isotopes. Subsequently, a Zircaloy cladding exhibiting a $17 \mu\text{m}$ oxide layer containing Li and B was analysed using a Cs^+ source and positive ions. It was found that due to its very high ionisation yield, Li can be easily detected by SIMS. Figure 1 shows colour coded ion maps for Zr and Li. Boron has a much lower ionisation yield than Li and consequently its analysis is more difficult. Nevertheless, it can still be detected.

At ITU, the SIMS will be used in conjunction with the shielded electron microprobe taking advantage of the

HIGHLIGHT

Thermophysical Characterisation of UO_2 Fuel with a Burn-up up to 100 MWd/kgHM

In 2004 the first measurements of the thermal diffusivity of highly irradiated commercial PWR fuel (burn-up $\sim 100 \text{MWd/kgHM}$) were performed using the shielded laser-flash apparatus, LAF-1 [1]. Unlike preceding measurements made on samples irradiated under isothermal conditions [2], power reactor fuel has to be examined locally due to the radial temperature gradient occurring during reactor irradiation, and the accompanying variation in the local microstructure and burn-up. However, the many cracks usually present in power reactor fuel makes it difficult to obtain a suitable sample for local laser-flash measurements along the complete fuel radius. These problems were successfully overcome in 2004 by optimisation of the sample preparation method in the hot cells and by a modification of the LAF-1 apparatus and improvement in experimental procedures.

Thermal diffusivity measurements were performed on 2 samples. The radial distribution of Nd in the UO_2 fuel matrix, as measured by electron probe microanalysis (EPMA), was used to obtain the local burn-up in the samples.

- For the first sample, thermal diffusivity measurements were performed at about the mid-radius position employing 6 consecutive thermal annealing cycles of constantly increasing maximum temperature up to 1100 K at which temperature the sample failed. The results for the first sample (see Fig. 1 HL) show a progressive recovery of the thermal diffusivity as a function of the maximum annealing temperature.
- For the second sample, thermal diffusivity measurements were performed at each annealing temperature at 6 radial positions. This was accomplished by moving the pyrometer spot by 1.0 mm dia between each measurement. The first position measured was in the vicinity of the pellet rim and the last position was almost in the centre of the pellet. The thermal diffusivity was first measured as a function of the pellet radius at 500K: the results demonstrate that it increases from the pellet rim to the pellet centre (see Fig. 2 HL). This dependence is linked to the decrease of the burn-up and to the increase of the fuel irradiation temperature. A first annealing cycle was then performed with measurements at temperatures 500, 600 and then again at 500K. This was followed by a second annealing cycle with measurements at

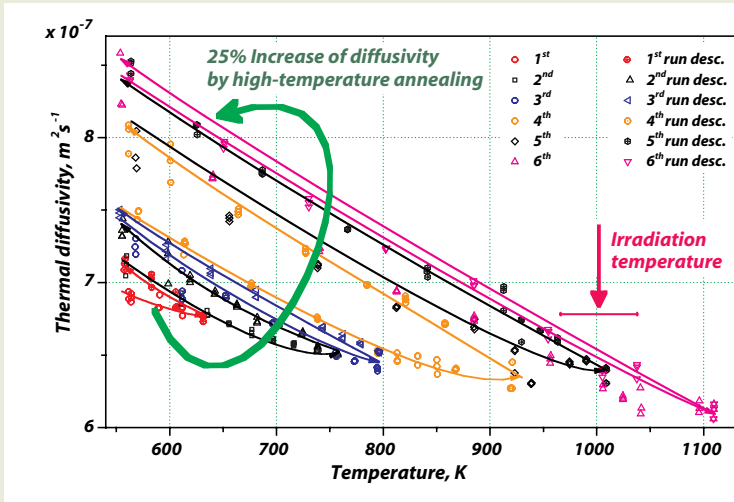


Fig. 1 HL: Thermal diffusivity of UO_2 fuel irradiated to 102 MWd/kgHM, measured at $r/R_0 = 0.55$ employing six consecutive thermal annealing cycles of constantly increasing maximum temperature.

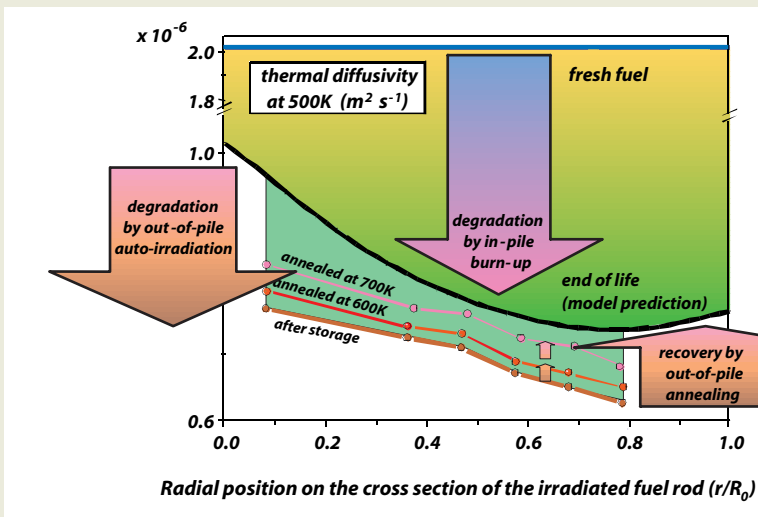


Fig. 2 HL: Thermal diffusivity at 500K measured in 6 radial positions after storage, after 2 annealing cycles and comparison with the end of life and fresh fuel values. 500K is the lowest temperature at which thermal diffusivity can be measured in the laser-flash device.

500, 700 and 500K. An increase, or recovery, in thermal diffusivity was observed after each annealing cycle. This recovery corresponds to the progressive annealing of the out-of-pile auto-irradiation damage (see Fig. 2 HL).

The thermal diffusivity values and the annealing behaviour measured on this highly irradiated commercial fuel is similar to that previously found for “research-grade” high burn-up fuels [2]. This analogy opens the perspective for further detailed investigation of various fuels homogeneously irradiated under controlled conditions, in which experimental investigations are easier and less time consuming to perform. These new results allow for the first time a precise assessment of the thermal diffusivity in the outer region of a power reactor fuel.

An important issue of thermo-mechanical behaviour of irradiated fuel is the role of the pressure constraint around

the fuel. When performing laser-flash measurements of thermal diffusivity of irradiated fuel [2] with no cladding, the fuel sample disintegrates close to the irradiation temperature. In the reactor however, a fuel is constrained and this probably raises the fuel fragmentation temperature. In order to simulate such effects and create a more versatile and flexible facility, ITU has undertaken to construct the POLARIS instrument. The 6 kW laser for this facility was delivered in 2004 and the first results are expected in 2005.

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3. Safety of Nuclear Fuels

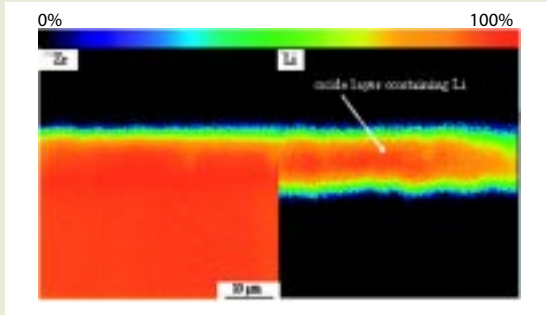


Fig. 1: False colour ion maps for the distribution of zirconium (left) and lithium (right) in a sample of Zircaloy cladding exhibiting a 17µm oxide layer. Resolution 256*256 pixels. Total dwell time 500 seconds. The lithium map clearly shows that lithium is located in the oxide layer.

complementary nature of the two techniques and is expected to make important contributions to ongoing research programmes

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3.2 HTR Fuel Testing

In the framework of a Share Cost Action (SCA) of the European Commission, a European Project aimed at the development of High Temperature Reactor (HTR) technology has been approved. The project includes developments in the fields of reactor physics, fuel technology, safety, material needs and the feasibility of key components and systems.

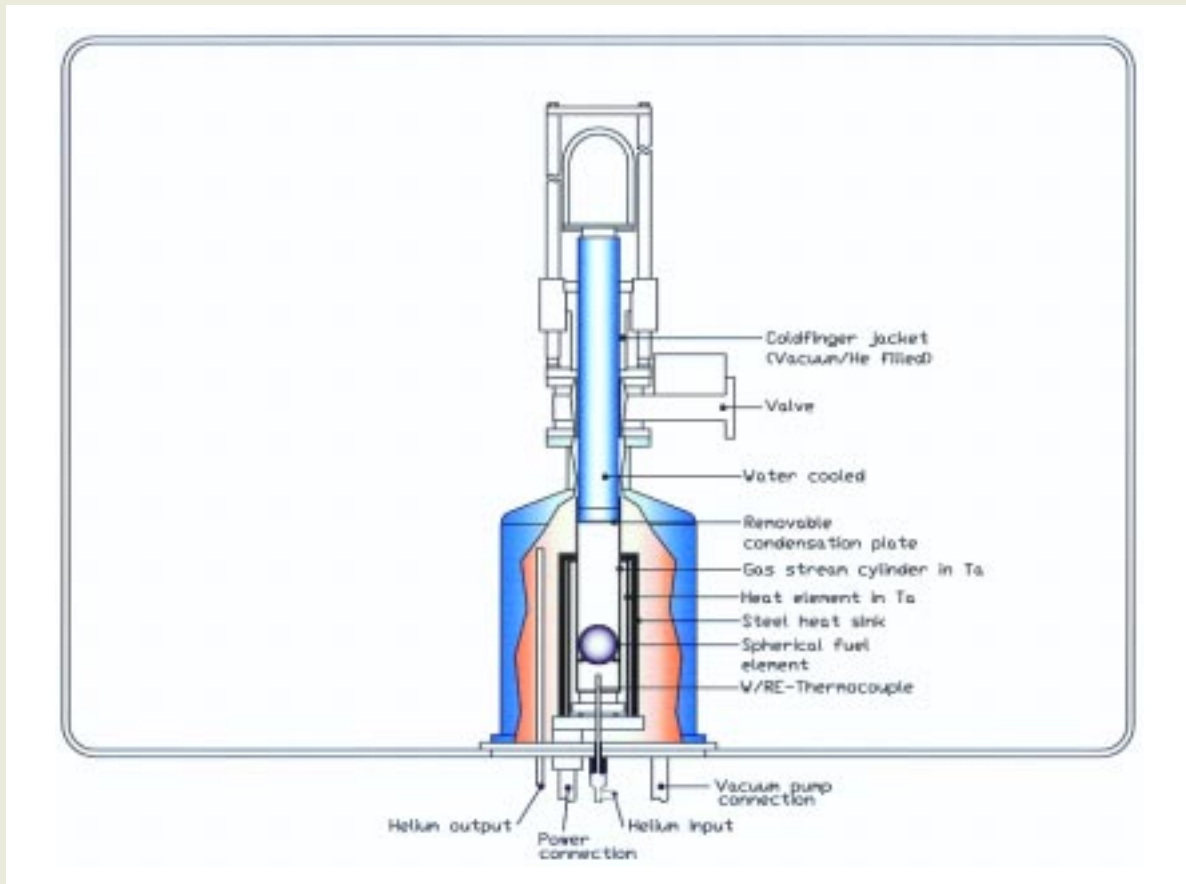


Fig. 2: The modified cold finger apparatus (KÜFA) for the measurement of fission product release from HTR fuel spheres

A key point in the domain of fuel technology is the testing of the irradiation behaviour of new fuel types and their fabrication methods. In this context, the testing of irradiated fuels under loss-of-coolant accident conditions will be needed to assess the quality of new fuel concepts and fabrication methods. That means, principally, the evaluation of the release behaviour of fission gases (Xe, Kr) and solid fission products (Cs, Sr, Ag, etc) under these off-normal conditions.

In the past, the so-called Cold Finger Apparatus (KÜFA) was developed in the Forschungszentrum Jülich (FZJ) to test HTR fuel design and fabrication methods. Using this device, the fission product release from defected particles can be tested up to 1800 °C.

In the framework of the SCA/HTR-technology, an upgraded version of the KÜFA (see Fig. 2) has been installed in the hot cells of the Institute for Transuranium Elements.

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3.3 Development of a Method for the Production of Inert Matrix Fuel Kernels for Plutonium Disposition in High Temperature Reactors (HTRs)

In a first step the sol gel internal or external gelation process is used to produce porous kernels of yttria stabilised zirconia (YSZ) in an inactive laboratory. These kernels are then entered in a glove box and are infiltrated with plutonium nitrate solution, before being calcined in air at 800°C to convert the nitrate to oxide. Thereafter, they can be sintered directly, or their plutonium content can be increased by additional infiltration and calcination cycles. The (Zr,Y,Pu)O₂ kernels shown in Fig. 3 were produced using such a two step infiltration process and had a Pu content of 11 wt%.

Dilution of the plutonium in YSZ not only permits the use of this essentially waste free production route, but also provides a material with an improved irradiation behaviour. For HTR applications, the first coating layer of a TRISO coated particle is 90 µm thick, and has a porosity of about 50%. Thus, the free volume in a coated particle

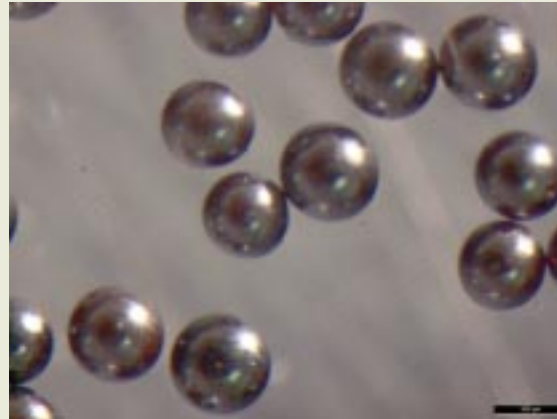


Fig. 3: (Zr,Y,Pu)O₂ kernels ($\varphi = 700 \mu\text{m}$) produced by a combination of the sol gel external gelation and Pu infiltration routes

in which the Pu is diluted to 10% in a 500 µm YSZ kernel is about five times greater than in the corresponding undiluted 200 µm PuO₂ kernel with the same Pu content. In this way, a larger volume is available to accommodate fission gas, and higher plutonium burnups can be achieved.

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3.4 Matrix Swelling Rate and Cavity Volume Balance of UO₂ Fuels at High Burn-up

The volume change of the active column of nuclear fuels during service in the reactor is a fact that was recognized in the early irradiation experiments. Because of its impact on heat transfer at the fuel-cladding gap (gap closure) and cladding strain, this issue has attracted the attention of designers and engineers over the years. At high burn-up the effect is dominated by the fuel expansion caused by precipitation of insoluble fission products, which in either solid or gaseous state require more space than the precursor U-atoms in the lattice [1]. Under normal PWR irradiation conditions, i.e., in the absence of thermally driven bubble growth, the phenomenon constitutes the so-called solid or inexorable swelling that depends only on burn-up and which cannot be suppressed or mitigated by application of exter-

3. Safety of Nuclear Fuels

nal pressure. Therefore, a critical evaluation of solid swelling has been performed up to burn-ups of 100 GWd/tM (100 MWd/kgHM), taking into account measured data for fuel density, porosity and retained Xe concentration by EPMA (Electron Probe Microanalysis). The work was completed with a theoretical assessment of the swelling contribution by fission gases under the realistic consideration of their fractions in dynamic solution and in intragranular and intergranular bubbles. This allowed proper conversion of the EPMA data into swelling data.

The results are summarized in Fig. 4, which shows the evolution with burn-up of all the terms constituting the basic volume balance equation of the fuel; namely,

$$\rho/\rho_{th} \cong 1 - \Delta V^{matrix}/V_0^{matrix} - P$$

where $\Delta V^{matrix}/V_0^{matrix}$ is the total inexorable swelling with both contributions of solid and gaseous fission products, ρ/ρ_{th} is the relative immersion density and P is the porosity. In Fig. 4 separate plots of the terms ρ/ρ_{th} and $1 - \Delta V^{matrix}/V_0^{matrix}$ as a function of burn-up are presented; the porosity, P, is obtained by difference. It is seen that up to burn-ups of ~60 GWd/tM the matrix swelling rate follows the slope ~1%/10 GWd/tM, which agrees with the literature data. However, in association with the formation of the high burn-up structure, at average pellet

burn-ups ≥ 60 GWd/tM the total matrix swelling rate gradually decreases from ~1 %/10 GWd/tM to ~0.3 %/10 GWd/tM; the latter being the typical contribution of solid fission products. The effect is due to the progressive depletion of fission gases in the fuel matrix by thermal and athermal processes, such that at very high burn-ups the matrix swelling becomes governed only by the precipitation of solid fission products. Owing to volume conservation, the volume occupied by the depleted gases is replaced by porosity.

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3.5 Further Development of the TRANSURANUS Code

The fuel modelling group is involved in a large set of projects dealing with UO₂ and MOX fuel for PWR, BWR, VVER and CANDU fuel. For 2004, two main achievements can be reported.

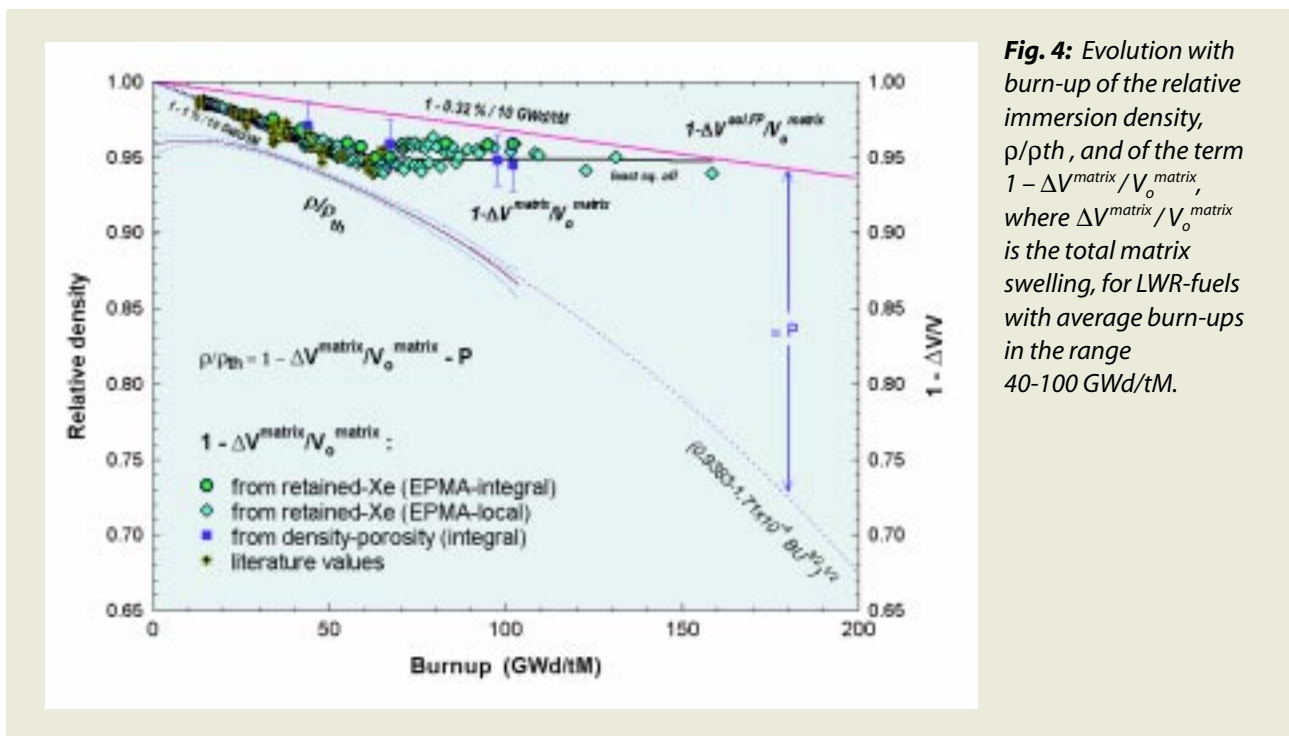


Fig. 4: Evolution with burn-up of the relative immersion density, ρ/ρ_{th} , and of the term $1 - \Delta V^{matrix}/V_0^{matrix}$, where $\Delta V^{matrix}/V_0^{matrix}$ is the total matrix swelling, for LWR-fuels with average burn-ups in the range 40-100 GWd/tM.

Modelling the behaviour of Zr1%Nb cladding under accident conditions

During 2004 a new effort was launched in collaboration with the Hungarian partners from AEKI Budapest and the German expert organization for nuclear fuel licensing (TÜV) for extending the code capabilities for VVER fuel rods under accident conditions. At AEKI un-irradiated cladding specimens were oxidised and loaded with hydrogen in a controlled, mixed hydrogen-steam atmosphere at different temperatures (900-1200°C) and different times. It was found that increasing the hydrogen content in the atmosphere considerably hinders the oxidation kinetics. The mechanical properties (yield stress, tensile strength, deformation) of the samples will be measured in 2005 by uniaxial, standardized ring compression tests, tensile tests and biaxial cladding ballooning tests. This will be complemented by metallographic examinations as well as hot extraction to determine the hydrogen content in the cladding. The data will be used to development a model for Zr1%Nb cladding oxidation and creep in the temperature range 900 to 1200°C along with a model for hydrogen uptake leading to new cladding failure criterion.

Improvement of the code predictions for MOX fuel

The version of the TRANSURANUS code for LWR MOX fuel is based on the standard version for LWR UO₂ fuel which takes into account the production of plutonium during irradiation. During 2004, specific attention was

devoted to the fuel thermal conductivity and its evolution with burnup, as well as the extended validation of the MOX version of the code in collaboration with ORNL in the USA. A new correlation for thermal conductivity of MOX fuel with burnup values between 20 and 45 MWd/kgHM [1] derived from laser flash measurements at ITU was compared with four alternatives from the open literature. An analysis of central temperature predictions as illustrated in Fig. 5 showed that the differences between measured and calculated values, and between the values calculated with the different options of the thermal conductivity are of the order of $\pm 5\%$. This is comparable to the experimental uncertainties on the power calibrations and the thermocouple calibrations. For all MOX fuel types and irradiation conditions covered in this work, the TRANSURANUS code performance can therefore be considered comparable to that achieved for standard LWR UO₂ fuels.

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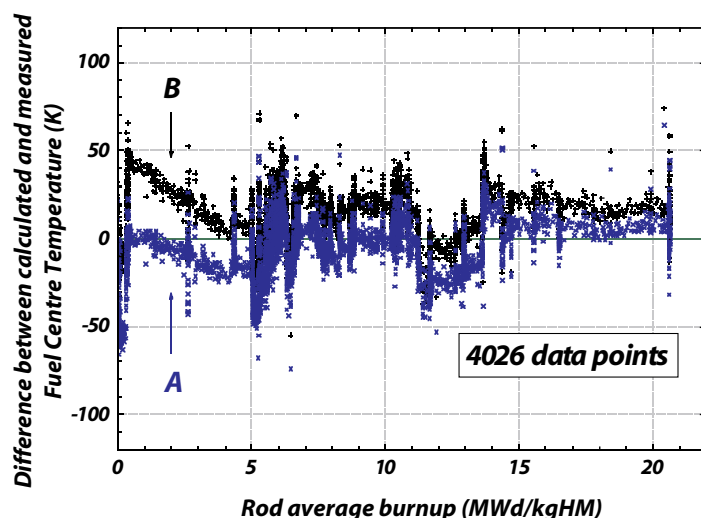


Fig. 5: Differences between in-pile measurements of fuel centre temperatures in a MOX rod irradiated at the OECD Halden Reactor Project and values calculated by TRANSURANUS. The calculations are performed for two correlations of the MOX fuel thermal conductivity: A): according to data from laser flash measurements at ITU, B): according to Carbajo et al. [2]. The fuel centre temperature in the steady-state phases of the irradiation was about 800°C.

3. Safety of Nuclear Fuels

3.6 PHEBUS PF Post Irradiation Examination (PIE) of the Degraded FPT2 Bundle

The FPT2 test of the international Phebus PF project examined the scenario when steam-starved conditions prevail during a severe accident (the Zircaloy reacts and consumes all the steam and results in an excess of hydrogen temporarily). These results are to be compared with the previous FPT1 test in which there was an adequate steam supply and the bundle degraded under more oxidising conditions.

ITU completed the bundle sectioning to obtain 8 x 2 cm thick discs from the key zones for optical macroscopy. Two discs were taken from the upper degraded bundle and one from the upper corium pool. Two discs were also taken from the lower section filled with debris below

the pool. An important aspect of this work was to verify to bundle tomography that had been carried out at Cadarache by IRSN before the bundle was shipped to ITU for PIE. The second activity was the selection and extraction of 6 samples (25 mm diameter) from the discs for optical microscopy. Furthermore, two samples were extracted from the corium pool and the lower debris for dissolution and complete chemical analysis by inductively coupled plasma-mass spectroscopy (ICP-MS).

Bundle tomography revealed that a large central cavity was apparent above the corium pool at approximately one-third bundle height. At the top there were remnants of distorted, degraded fuel rods, whereas below the corium pool there was small streams of melt material and debris evident. In comparison with the previous test FPT1 the extent of degradation appears to be greater with a larger cavity and a deeper corium pool.

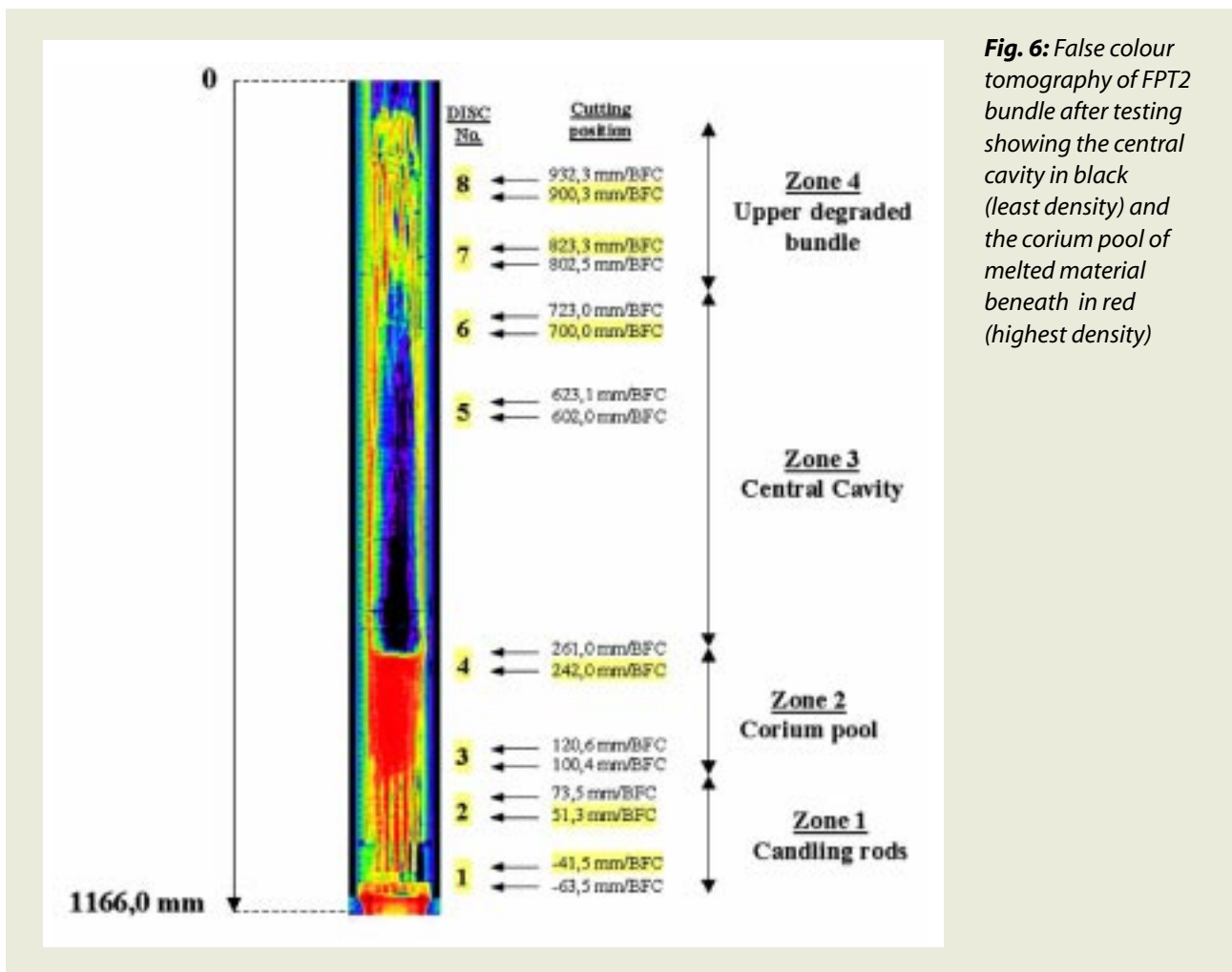


Fig. 6: False colour tomography of FPT2 bundle after testing showing the central cavity in black (least density) and the corium pool of melted material beneath in red (highest density)

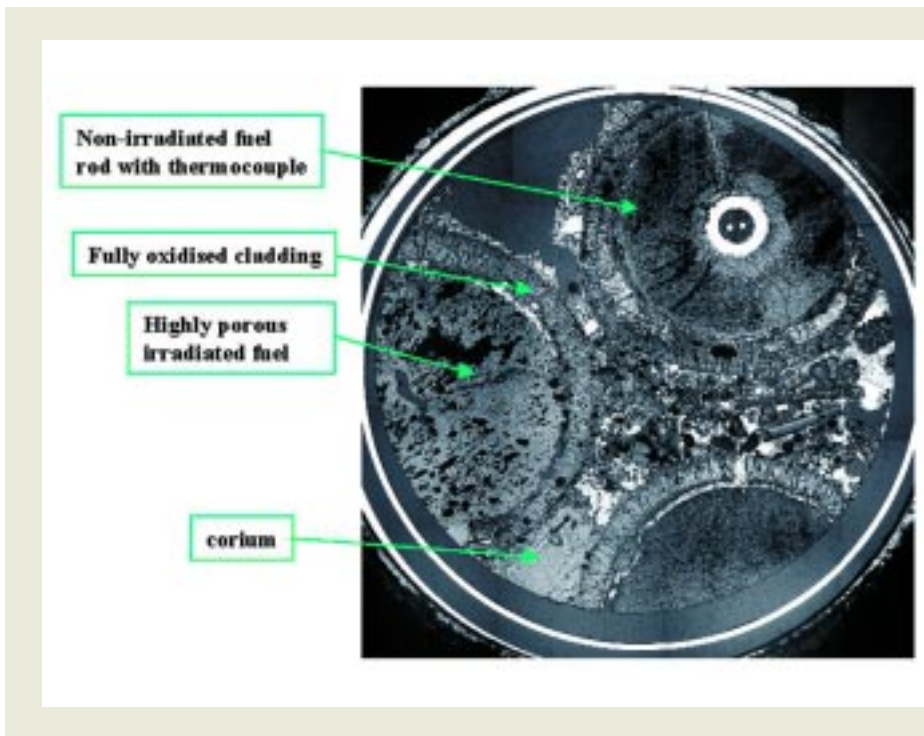


Fig. 7: Microscopic sample from Disc 7 of the upper degraded bundle. The highly porous irradiated fuel is seen along with thick fully oxidised cladding. Only a small amount of corium is present.

The main results of the bundle sectioning were to once again confirm the accuracy of the tomography carried out at IRSN, Cadarache. This confirmation was also required because the bundle appeared to have been displaced during the filling (embedding) of the bundle with a Wood's metal (a lead-bismuth-indium-cadmium mixture that melts at 70-120°C).

The upper part of the fuel rods appeared to have been considerably degraded and the cladding oxidised. There was little evidence of melt material in this upper region and this may have already relocated to the corium pool or even lower. The corium attack of the surrounding thermal protective shroud had just initiated.

The differences between the degraded bundle geometries of FPT1 and FPT2 can be explained by the fact that under steam-starved conditions (FPT2) Zircaloy metal melts and relocates at a lower temperature, whereas under oxidising conditions (FPT1) the Zircaloy cladding oxidises to a refractory oxide (ZrO_2) and remains in place until very high temperatures are reached later in the accident.

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4. Partitioning and Transmutation

4.1 Partitioning

Aqueous reprocessing

New centrifugal extractors (Robatel, France) have been installed in the hot cell facilities. The centrifuges have been adjusted and optimised for hot cell work and undergone a thorough cold testing in order to determine the performance and capacity. The preparation involved the manufacturing of a support structure for 16 centrifuges and 5 pumps, see Fig. 1. All parts of the centrifugal extractor set up in contact with solutions are made of titanium, which makes the system compatible with most solvent and acids (except hydrofluoric acid and high concentrations of sulphuric acid). The centrifugal system is flexible and allows different flow sheets to be built up and tested.



Fig. 1: Set up of the 16 centrifugal extractors installed in the hot cell

Several cold tests have been carried out leading to the following conclusions:

- Flexible system (any flow-sheet possible)
- Reliable (no failure during extensive testing). Total capacity of at least 2400 mL/h at organic/aqueous flow ratios =1 (compared to 400 mL/h of the old ones), O/A ratios from 0.1 up to 10.
- Flow-sheets carried out in old centrifugal extractor system (lower flow-rates) successfully tested.
- Equilibrium reached after 1 hour (DIAMEX extraction process, Feed 50 mL/h, Org. 75 mL/h).

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HIGHLIGHT

First Americium Fuel Fabrication in the Minor Actinide Laboratory

In recent years, different concepts have been proposed for fuels and targets for the transmutation of minor actinides. They can be categorised in terms of fuel material (metal, ceramic), fuel form (homogeneous, composites) and fuel packing (pellet, particle, liquid salt). In all cases, however, the transmutation efficiency must be maximised, a condition best achieved if uranium-free fuels are considered. The choice of the inert matrix for uranium-free fuels is a key aspect of this research [1]. The actinide phase and the inert matrix can be combined either in a homogeneous fuel form, in which the actinides form a solid solution with the matrix [2], or in a composite fuel form, in which the actinides are geometrically dispersed in a matrix [3]. Both of these fuel types have their advantages and disadvantages; e.g. the fabrication of the homogeneous fuel form is a well known process but such fuel forms usually have a low thermal conductivity. In the composites, on the other hand, the thermal properties are improved. In addition, the irradiation-induced property changes in the composite pellets are potentially minimised by localising the fission damage in a limited geometric domain within the fuel. Their fabrication, however, is considerably more difficult as a result of the specific requirements of size and homogeneous distribution of the dispersed actinide phase.

At ITU both fuel/target forms are being studied mainly for pellet-type compaction. Given the high radioactivity of the minor actinides, much emphasis is given to the development of clean and, necessarily, dust-free fabrication methods to minimize contamination in the gloveboxes. A hybrid process consisting of a combination of sol gel and porous bead infiltration techniques was developed for the fabrication on a semi-industrial scale. It meets three critical criteria: (a) it is dust free, (b) there is no liquid waste, and (c) it minimises the steps in which highly radioactive materials are handled. In July 2004 the start of the active operation of the minor actinide laboratory marked a significant milestone for actinide fuel R&D at ITU.

Homogeneous pellets of $\text{Am}_{0.06}\text{Y}_{0.16}\text{Zr}_{0.78}\text{O}_{2-x}$ were first to be fabricated. In a first step highly porous beads of an yttria-stabilised zirconium oxide $\text{Y}_{0.17}\text{Zr}_{0.83}\text{O}_{1.92}$ (YSZ) were produced by the external gelation method, which guarantees a solid solution as the final product. These YSZ beads have a polydisperse size distribution in the 40 to 150 μm range, a specific surface area of $67 \text{ m}^2 \cdot \text{g}^{-1}$ and their porosity is about 80%. Prior to their infiltration, the YSZ beads were placed in a quartz container and the americium nitrate solution ($120 \text{ gAm} \cdot \text{l}^{-1}$) was added dropwise until the incipient wetness point was reached. At this point all the beads were completely infiltrated and there was no excess liquid, which could cause agglomeration or caking of the beads in the later stages of the process. After infiltration, the beads were calcined at 800°C for 2 hours under an air atmosphere to convert the infiltrated metal nitrate to oxide. The americium content was derived from gravimetric measurements of the beads before their infiltration and after conversion of the infiltrant nitrate to oxide by calcination.

Following calcination, the resulting infiltrated particles were free-flowing, and, due to their size and integrity, their physical manipulation does not produce dust in the remaining fabrication steps. The beads were pressed into pellets and then thermally treated at 1000°C in Ar/H_2 followed by sintering at 1650°C in Ar . The final pellets have a density of $94.5 \pm 1.0\%$ theoretical density (TD), calculated from geometrical and gravimetric measurements with referenced to $\text{TD} = 6.4 \text{ g} \cdot \text{cm}^{-3}$. Visual inspection of the surface of the pellets shows that they have a perfect cylindrical geometry and excellent integrity without macro or micro cracks (see Fig. 1 HL).

In addition, $\text{Am}_{0.20}\text{Y}_{0.16}\text{Zr}_{0.64}\text{O}_{2-x}$ -MgO (ceramic-ceramic) pellets were also fabricated. After their calcination the YSZ beads were sieved and specific size fractions (40-60 and 100-125 μm) were selected. Each size fraction was infiltrated with an americium nitrate solution ($269 \text{ g Am} \cdot \text{l}^{-1}$). Two infiltration-calcination steps were required to reach the desired metal content, i.e. $\text{Am}/(\text{Am}+\text{Y}+\text{Zr}) = 0.20$. The $\text{Am}_{0.20}\text{Y}_{0.16}\text{Zr}_{0.64}\text{O}_{2-x}$ phase was mixed with the required amount of MgO (CERAC M-1017) and, following addition of zinc stearate as lubricant, the mixtures were compacted in-

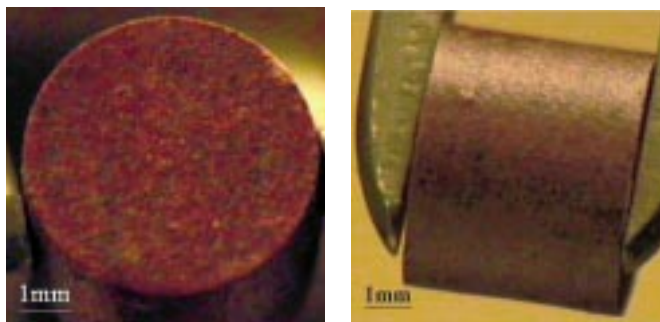


Fig. 1 HL: Homogeneous $\text{Am}_{0.06}\text{Y}_{0.16}\text{Zr}_{0.78}\text{O}_{2-x}$ pellet

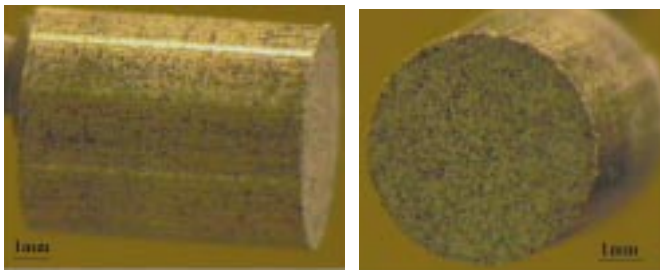


Fig. 2 HL: Ceramic-ceramic (CERCER) $\text{Am}_{0.20}\text{Y}_{0.16}\text{Zr}_{0.64}\text{O}_{2-x}$ -MgO composite

to pellets. Then they were thermally treated in Ar/H_2 followed by sintering in Ar . The final pellets have a density of $90.0 \pm 1.0\%$ TD. Visual inspection of the surface of the pellets produced, show that they have a perfect cylindrical geometry, excellent integrity and with a random distribution of isolated beads, regardless of the sphere size (see Fig. 2 HL).

Three fuel pins fabricated at ITU, one homogeneous and two composite fuel type, all containing $0.7 \text{ g} \cdot \text{cm}^{-3}$ americium, will be irradiated in the fast neutron flux in Phenix in 2006¹.

References

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¹ These activities are performed under a CEA contract.

4. Partitioning and Transmutation



Fig. 2: Al electrode covered with 1.13 g of actinides (a) and the same electrode after treatment in HNO_3 (b)

Pyrochemical reprocessing

New separation methods for actinides by electrolysis in molten salt media, so called pyro-processing, are being researched for advanced fuel cycles. These studies are concentrated on electrorefining of metallic $\text{U}_{60}\text{Pu}_{20}\text{Am}_2\text{Ln}_5\text{Zr}_{10}$ fuel in LiCl-KCl eutectic. In these separation experiments, pieces of metallic alloy are loaded into an anode basket and dipped into the molten salt containing actinides and lanthanides salts. Electrorefining is then carried out by applying a constant current between the basket and an aluminium cathode. This causes the metallic alloy fuel to anodically dissolve, feeding U^{3+} ,

Pu^{3+} , Am^{3+} and Ln^{3+} to the salt. On the cathode actinides are collected together onto the Al foil as An-Al alloys leaving lanthanides in the salt phase.

In total more than 20 such experiments have been carried out involving approximately 25 g of metallic alloy fuel. Figure 2 shows pictures of one of the cathodes after electrodeposition (see Fig. 2, a) and after dissolution with nitric acid (see Fig. 2, b). In the treatment with nitric acid, the reaction with the An-Al alloy deposit seemed to be complete and no significant dissolution of the pure Al electrode occurred. This is probably due to a passivation of the Al surface by an oxide layer. An expected Al/An ratio in the dissolution solution of at least 4 is expected since An are deposited as AnAl_4 . The dissolution solutions were analysed by ICP-MS and the composition of the deposits is listed in Table 1.

	Mass in the deposit (mg)		
	Electrolysis 1	Electrolysis 2	Electrolysis 3
Cathodic potential (V)	-1.20 to -1.27	-1.20 to -1.35	-1.20 to -1.25
U	512	151	971
Pu	243	137	433
Am	13.5	11.5	24.1
Zr	–	4.19	7.08
Y	0.018	0.023	0.076
Ce	0.026	0.035	0.043
Nd	0.244	0.340	0.437
Gd	0.025	0.038	0.061
$m_{\text{An}} / m_{\text{Ln}}$	2451	687	2314
$m_{\text{Am}} / m_{\text{Ln}}$	43.0	26.4	39.1

Table 1: Composition of the cathodes

From Table 1, it can be seen that the deposits are composed mainly of U, Pu and Am and that they contain only small amounts of lanthanides. The highest An/Ln mass ratio (~2400) is obtained when the cathodic potential is kept more positive than -1.3 V (electrolysis 1 and 3). This demonstrates that this process can achieve a very efficient separation of An from Ln. The Am/Nd con-

centration ratio in the starting salt solution (0.40) has been increased approximately 100 times to a Am/Ln ratio of about 40 in the deposit (electrolysis 1 and 3). In the second electrolysis the cathodic potential was more negative, which can explain the lower Am/Ln ratio of 26, as more lanthanides are reduced with decreasing potential. Despite the low concentration of Am compared to Ln in the salt phase, an efficient separation of MA from Ln has been demonstrated.

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4.2 Studies of Advanced Nuclear Processes

Laser Transmutation Studies

Laser accelerated electrons generated in a He gas jet, were targeted onto high-Z materials for conversion to high-energy bremsstrahlung [1]. Nuclear reactions in targets were analyzed with time-resolved γ -spectroscopy. The electron-jet temperature of more than 35 MeV, enabled us to efficiently generate photo-induced nuclear reactions. In addition to confirming our previous cross section measurements on ^{129}I , the photo-induced transmutation yields were increased by two orders of magnitude compared to earlier experiments.

Laser accelerated protons were used for the first time to investigate experimentally residual nuclide production in lead spallation targets [2]. The potential of high power laser technology for the measurement of data relevant to accelerator driven systems was demonstrated.

Neutron production rates using laser accelerated protons were investigated [3]. With the VULCAN giant pulse laser, more than 10^9 neutrons per shot were produced. In contrast, existing table-top lasers can produce 10^6 - 10^7 neutrons per second. Next generation table-top lasers, currently under construction, will be capable of producing nanosecond neutron pulses at a rate of 10^{10} neutrons per second. The short pulse length and strength of such neutron sources is of interest for pulsed fast neutron activation (PFNA) for 3D spatial distribution detection of explosives, drugs and nuclear materials in full-sized shipping containers.

The above work was done in collaboration with the Institute for Quantumelectronics, University of Jena (Ger-

many), University of Strathclyde (UK), Rutherford Appleton Laboratory (UK), and the Jožef Stefan Institute (Slovenija).

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- [2] P. McKenna et al., Phys. Rev. Lett. 94 (2005) 084801
- [3] T. Žagar et al., to be published in 2005

A new book: "Radioactivity – Radionuclides – Radiation" was published by Springer Verlag (for information see www.nuclides.net). This book has arisen out of various activities at ITU (nuclear forensics, laser transmutation, environmental radioactivity, etc.) including a series of training courses held over the past two years. Radioactivity – Radionuclides – Radiation is suitable for a general audience interested in topical environmental and human health radiological issues such as radiation exposure in aircraft, food sterilisation, nuclear medicine, radon gas, radiation dispersion devices, etc. It leads the interested reader through the three Rs of nuclear science, to the forefront of research and developments in the field. The book is also suitable for students and professionals in the related disciplines of nuclear and radiochemistry, health physics, environmental sciences, nuclear and astrophysics. Recent developments in the areas of exotic decay modes (bound beta decay of 'bare' or fully ionized nuclei), laser transmutation, nuclear forensics, radiation hormesis and the LNT hypothesis are covered.

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4.3 Investigation of the Thermodynamic and Vibrational Properties of Ceramics for the Immobilisation of Actinides

The suitability of the pseudo-ternary system $(\text{Ln},\text{An})\text{PO}_4\text{-M}^{\text{II}}(\text{M}^{\text{IV}},\text{An})(\text{PO}_4)_2\text{-AnSiO}_4$ (with Ln = La or Ce, M^{II} = Ca or Ba and M^{IV} = Zr or Hf) as a matrix for the im-

4. Partitioning and Transmutation

mobilization of both trivalent and tetravalent actinides (An) is currently being investigated. The research activities are focussed on the preparation of the possible end-members of this system, the study of the phase relationships (miscibility) and the testing of their aqueous stability. This work is done in close collaboration with the Institute for Mineralogy of the University of Münster (Germany).

During 2004 LaPO_4 , CePO_4 , PuPO_4 , $\text{BaZr}(\text{PO}_4)_2$ and $\text{BaHf}(\text{PO}_4)$ end-member compounds of the binary (see above) subsystem $(\text{Ln}, \text{An})\text{PO}_4\text{-M}^{\text{II}}(\text{M}^{\text{IV}}, \text{An})(\text{PO}_4)_2$ were synthesised and characterised. PuPO_4 was synthesised by precipitation from a Pu-nitrate solution by adding Na_2HPO_4 in stoichiometric amounts. The precipitate was thermally treated at $T = 873 \text{ K}$ (in Ar) and $T = 1273 \text{ K}$ (in Ar/ H_2). It was characterised by X-ray diffraction (XRD) and its low-temperature heat capacity was measured. $\text{BaZr}(\text{PO}_4)_2$ was synthesised from equimolar quantities of BaCO_3 , ZrO_2 and $(\text{NH}_4)_2\text{HPO}_4$. The ceramic was investigated by high-temperature XRD, DSC, and drop calorimetry. A previously unknown phase transition from a monoclinic to a hexagonal structure was detected by all techniques.

Some pseudo-binary subsystems were also studied. In the $\text{LaPO}_4\text{-BaZr}(\text{PO}_4)_2$ system a limited miscibility of up to 20 mol% $\text{BaZr}(\text{PO}_4)_2$ was found. The mixtures of $\text{CePO}_4\text{-PuPO}_4$ showed a mutual miscibility but a second-phase was formed. This will be investigated further.

The solid solution $(\text{La}_{1-x}\text{Eu}_x)\text{PO}_4$ with $0 < x < 1$ was studied in more detail as a proxy of the $\text{LnPO}_4\text{-AnPO}_4$ system. The solid solutions were analysed by XRD, Raman and infrared (IR) spectroscopy. Whereas precise XRD measurements allowed detecting an excess volume, vibrational spectroscopy was used to characterize strain fields associated with the substitution of one cation by another. Such strain fields are reflected by line broadening due to an overlap of length scales for phonons and any local elastic heterogeneity and are believed to be the main cause of an excess enthalpy in regular solid solutions. In ideal solid solution the excess enthalpy will be close to zero and the aqueous solubility will be a linear function of the end member solubility. In contrast, regular solid solutions are characterized by internal strain and a positive excess enthalpy and the intermediate solid solutions may have a higher solubility compared to the end members. A detailed analysis of the Raman and IR spectra of the $(\text{La}, \text{Eu})\text{PO}_4$ solid solution indicates that the intermediate compounds show significant stronger line broad-

ening than the end members (see Fig. 3), likely reflecting an excess enthalpy. Future work will concentrate on studying the dependence of the hydrothermal stability on such elastic strain, i.e. on the thermodynamic properties of the solid solution. First hydrothermal alteration tests at 300°C indicate that the solid solution is nevertheless extremely stable over the whole compositional range.

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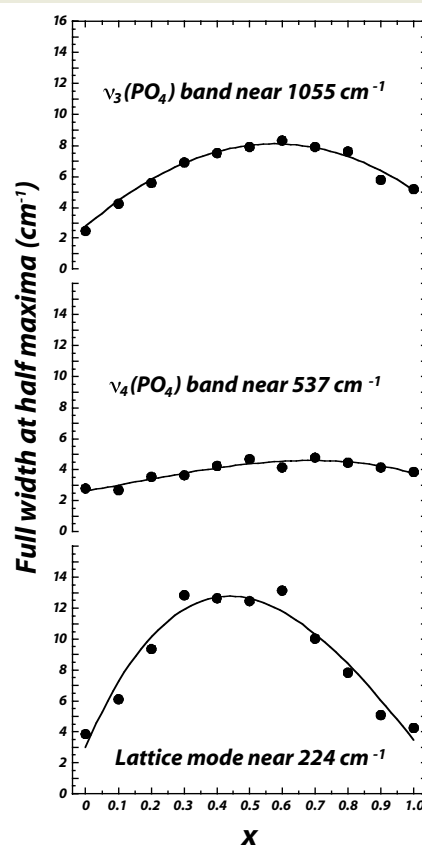


Fig. 3: The line width expressed as the full width at half maximum (Γ) of three representative Raman modes as a function of x in the $(\text{La}_{1-x}\text{Eu}_x)\text{PO}_4$ solid solution. Solid lines represent best fits with an equation similar to the asymmetric functions for excess properties of subregular solid solution models, i.e.

$$\Gamma = a_0 + a_1x + x(1-x)[a_2(1-x) + a_3x]$$

Errors of the line width are roughly within the size of the symbols.

Selected Publications

B. Christiansen, C. Apostolidis, O. Courson, R. Malmbeck, R. Carlos, G. Pagliosa, K. Römer, D. Serrano-Purroy, J.P. Glatz

Advanced aqueous reprocessing in P&T strategies: process demonstrations on genuine fuels and targets. *Radiochim. Acta* 92 (2004) 475

M. Kurata, T. Inoue, J. Serp, M. Ougier, J.P. Glatz
Electro-chemical reduction of MOX in LiCl. *J. Nucl. Mater.* 328 (2004) 97

B. Liesfeld, K.W. Amthor, F. Ewald, H. Schwoerer, J. Magill, J. Galy, R. Schenkel, R. Sauerbrey
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J. Serp, R.J.M. Konings, R. Malmbeck, J. Rebizant, C. Scheppler, J.P. Glatz
Electrochemical behaviour of plutonium ion in LiCl-KCl eutectic melts. *J. Electroanal. Chem.* 561 (2004) 143

Collaborations

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5. Alpha-Immunotherapy/ Radiobiology

The alpha-immunotherapy group has continued to contribute to the development of drugs for targeted alpha therapy (TAT) of various cancers and infectious diseases. ITU mainly focuses on the production of radioisotopes, the chelation of carrier molecules and radiobiological studies on radiation-induced effects on cells. Additionally, ITU supports numerous pre-clinical and clinical studies in hospitals and cancer research centres throughout Europe and worldwide to demonstrate the feasibility of targeted alpha therapy. An overview of ITU's current collaborations in the field of TAT is given in Table 1.

5.1 Production of Radionuclides for Targeted Alpha Therapy: $^{225}\text{Ac}/^{213}\text{Bi}$

The production of alpha emitters remains a key prerequisite for the development of targeted alpha therapy. Only ITU and Oak Ridge National Laboratory (USA) can presently provide $^{225}\text{Ac}/^{213}\text{Bi}$, the alpha-emitters mainly used today. In 2004, 31 $^{225}\text{Ac}/^{213}\text{Bi}$ -generators, corresponding to 554 mCi in total, and 2 shipments of ^{90}Y (29 mCi in total) were provided by ITU to our various partners. Stringent quality control of the final product ensured reliable functioning of the generators provided by ITU.

5.2 Pre-Clinical and Clinical Studies Supported by ITU

In the framework of collaborations with hospitals and cancer research centers, ITU has continued to support pre-clinical and clinical studies by providing radionuclides, training and know-how, specific equipment necessary for the handling of alpha-emitters and by chelation of carrier molecules. In 2004, studies on the application of TAT for the treatment of various types of cancers and infectious diseases were continued *in vitro*, in animals or within clinical trials in patients.

The evaluation of the *in vitro* response caused by TAT in comparison with different cytotoxic treatments was continued at the University of Ghent. The studies showed that the induction of apoptosis by conventional therapies for SLVL (splenic lymphoma with villous lymphocytes) and B-CLL (chronic lymphocytic leukaemia) patients did not differ significantly for fludarabine, methylprednisolone and external gamma irradiation, but apoptotic scores were significantly higher after treatment with the anti-CD20 an-

HIGHLIGHT

From Bench to Bedside: Targeted Alpha Therapy (TAT) for Malignant Melanoma

Collaboration between ITU and the Centre for Experimental Radiation Oncology, Sydney

One of ITU's long-standing cooperation partners is the Centre for Experimental Radiation Oncology (CERO) at St. George Hospital Cancer Care Centre in Sydney, Australia (Head: Prof. Barry J. Allen). As in previous years, ITU continued in 2004 to ship ^{225}Ac generators to CERO. The 4 shipments contained in total approx. 50 mCi of ^{225}Ac and 29 mCi of ^{90}Y and allowed numerous studies of pre-clinical alpha and beta therapy and a phase I clinical trial of TAT for malignant melanoma. This is a skin cancer that has its highest incidence in Australia and while surgery for early stage disease can be successful, there is no systemic therapy available to control metastatic melanoma.

CERO is currently concentrating on using ^{213}Bi conjugated with the monoclonal antibody (mAb) 9.2.27 directed against the human melanoma proteoglycan NG2. The clinical applications address stage IV melanoma patients with secondary subcutaneous melanoma, high-risk patients after resection of primary tumour. A phase I study aiming to establish the maximum tolerated dose of ^{213}Bi -cDTPA-9.2.27 mAb using a dose escalation design has been conducted at CERO. 16 patients received intratumoral alpha-immunoconjugate (AIC) injections with activities from 0.05 mCi up to 0.5 mCi, increasing in steps of 100 μCi . There were no adverse events except for momentary pain at the injection site. Levels of the melanoma serum marker MIA in treated patients were compared at baseline, 2 and 4 weeks post-TAT. A significant decrease at 2 weeks was observed in 4/6 patients, followed by an increase at 4 weeks. Immuno-histochemistry of tumour sections (see Fig.1 HL) showed that all melanomas were positive for 9.2.27 mAb and the AIC was locally efficacious at a dose above 0.15 mCi. Taken together, the reduced MIA levels, the immunohistochemistry results of the excised tumour, the apoptosis and Ki67 results indicate the melanoma response to therapy. Intralesional TAT was very effective in delivering a high radiation dose to the tumour and reducing the MIA levels. Most tumour cells

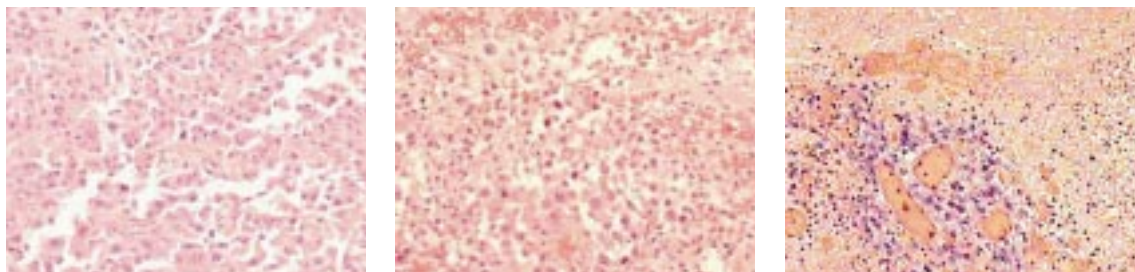


Fig. 1 HL: Stained sections of untreated (left), cold-antibody treated (middle) and ^{213}Bi -labeled 9.2.27-mAb treated (right) melanoma tumours. Untreated and cold-Ab treated sections show conventional histology, treatment with ^{213}Bi -labeled 9.2.27-mAb results in extensive cell debris (from [1]).

have been targeted and suffered apoptotic cell death. TAT may therefore delay the progression of melanoma.

Enlarging the spectrum of TAT applications, the $^{225}\text{Ac}/^{213}\text{Bi}$ generators provided by ITU were used for the production of new alpha-immunoconjugates designed to target pancreatic and ovarian cancer. Following the *in vitro* evaluation of the cytotoxicity of ^{213}Bi -labelled carrier molecules PAI2 and C595, their cytotoxicity was assessed *in vivo* in a xenograft model. Nude mice were inoculated with human pancreatic cancer cells. At 2 days post-inoculation, mice were injected with AIC by local or systemic injection. A single local subcutaneous (sc) injection of 150 MBq/kg could completely inhibit tumour growth and an intraperitoneal (ip) injection of 111 MBq/kg caused significant tumor growth delay. These results indicate that both ^{213}Bi -PAI2 and ^{213}Bi -C595 may be useful agents for the treatment of micro-metastatic pancreatic cancer with overexpression of uPA or MUC1 antigens in post-surgical patients with minimal residual disease.

ITU provided not only alpha-emitters. A new field of cooperation was developed around pre-clinical applications of beta-emitters. Two shipments of ^{90}Y (29 mCi) allowed to start investigation of animal survival after treatment with ^{213}Bi -immunoconjugates in comparison with ^{90}Y -immunoconjugates in a nude mice model using the targeting vector PAI2 which recognizes ovarian cancer cells. The relative biological effective (RBE) dose was calculated considering $\text{RBE}=4$ for alpha particles. Three treatment arms were investigated: alpha-treatment, beta-treatment and beta followed by alpha, with activities of 6 mCi (alpha), 10 mCi (beta), and 5 mCi (beta) followed by 3 mCi (alpha). The

study was successful in showing that all therapy groups resulted in superior survival compared with the untreated animals and in establishing effective doses for survival for ^{213}Bi , ^{90}Y and their combination. The results also indicated a value for $\text{RBE} (^{213}\text{Bi}/^{90}\text{Y}) > 1.7$.

A highlight of the year was the award of an Australian Government Innovation Grant that allowed the whole research group to travel to Germany for collaborative studies between ITU and CERO on TAT for lymphoma and to participate at the 4th Alpha-Immunotherapy Symposium in Düsseldorf. Four papers were presented, Prof. Allen chaired a session and gave the conference overview.

In accordance with the agreement between ITU and CERO, several initiatives were taken in the intellectual property area. CERO had lodged a provisional patent in Australia on the application of ^{213}Bi in TAT and ITU has followed up with a full patent application. The collaboration with the Australian colleagues has become a tradition for ITU's alpha-immunotherapy group. As we plan to continue and develop it, one of the CERO's staff members will join ITU in 2005 as a visiting scientist.

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5. Alpha-Immunotherapy/ Radiobiology

Partner	Cancer type or collaboration project
Technische Universität München, Nuklearmedizinische Klinik (D)	Gastric cancer
University of Düsseldorf, Department of Hematology, Oncology and Clinical Immunology (D)	Non-Hodgkin's lymphoma (NHL)
German Cancer Research Center (DKFZ) (D)	Non-Hodgkin's lymphoma (NHL)
University of Ghent, Department of Hematology (B)	Non-Hodgkin's lymphoma (NHL)
Virga Jesse Hospital, Hasselt (B)	Non-Hodgkin's lymphoma (NHL)
INSERM, Nantes (F)	Multiple myeloma
Clinic of Neurosurgery, Kantonsspital Basel (CH)	Glioma, glioblastoma
St. George Cancer Care Center & Medical Scitec (Australia)	Melanoma and prostate, pancreas, breast, ovarian cancer
Memorial Sloan-Kettering Cancer Center (USA)	Acute myelogenous leukemia (AML); Prostate cancer
Albert Einstein College of Medicine (USA)	Antimicrobial therapy
National Cancer Institute, Bethesda (USA)	Chelation Chemistry
JRC, Institute for Health and Consumer Protection, Ispra (Italy)	Radionuclide production
Nuclear Physics Institute, Rez near Prague (Czech Republic)	Radionuclide production

Table 1

tibody (rituximab) conjugated or not to ^{213}Bi , making SLVL patients potentially good candidates for TAT.

In collaboration with ITU, TAT of peritoneal carcinomatosis in diffuse-type gastric cancer was continued to be studied at the Technical University of Munich. Tumour cell dissemination into the peritoneal cavity after resection of a primary solid tumor often causes tumor recurrence. Studies comparing the therapeutic efficacy of single versus double intraperitoneal injection of ^{213}Bi -labelled d9Mab in a mouse model showed that repeated application of ^{213}Bi -d9Mab significantly prolonged mean survival in comparison to a single injection even at advanced stage of the disease.

In the course of the clinical phase I study "Alpha-Immunotherapy with a ^{213}Bi -coupled, chimeric anti-CD20 antibody (MabThera®) for patients with refractory B-cell malignancies" nine patients had been treated in past years (German Cancer Research Center, Universities of Heidelberg and Düsseldorf). The clinical trial was continued in 2004 at the University Hospital Düsseldorf

based on extensive support from ITU through provision of equipment and training of personnel. The trial is designed as dose escalation study to determine the tolerability of the treatment approach. Out of the first 3 patients treated, 1 patient received 13 mCi ^{213}Bi -labelled anti-CD20 antibody, 2 patients were treated on dose levels of 0.75 mCi/kg body weight, corresponding to 52 mCi and 58 mCi ^{213}Bi , respectively. In all 3 patients minor hematological toxicities were observed. Depending on tumor burden, the bone marrow seems to be the critical organ for therapy with ^{213}Bi -antiCD20. The study will be continued in 2005.

As in previous years, the major part of $^{225}\text{Ac}/^{213}\text{Bi}$ produced at ITU in 2004 was sent to Memorial Sloan-Kettering Cancer Center, New York to continue the study of TAT for acute myeloid leukaemia. In the ongoing phase I/II trial 26 AML patients were treated with standard dose cytarabine followed by ^{213}Bi -HuM195 to determine the effects of ^{213}Bi -HuM195 after partial cytoreduction with chemotherapy. During the phase I part of the study, the dose of ^{213}Bi -HuM195 was escalated from 0.5 to 1.25

mCi/kg. The maximum tolerated dose was determined to be 1 mCi/kg. No responses were seen at the first two dose levels. Seven of the 20 patients (35%) who received either 1 mCi/kg or 1.25 mCi/kg responded. There were 2 CRs (complete remissions) lasting 9 and 12 months; 3 CRp (CR with incomplete platelet recovery) lasting 1, 2, and 6 months; and 2 PRs lasting 3 and 8 months. The most common extramedullary toxicities were transient low-grade elevations in liver function tests and serum creatinine, and grade 1 or 2 infusion-related toxicities. The study shows that sequential administration of cytarabine and ^{213}Bi -HuM195 is tolerable and can produce complete remissions in patients with AML. In 2004 seven patients were treated at MSKCC, the study will be continued in 2005.

In the frame of a collaboration with Albert Einstein College of Medicine, New York, studies of TAT for the treatment of infectious diseases, including fungal and bacterial infections, were continued. The conventional treatment of infections is often hampered by increasing drug resistance rendering TAT a promising alternative approach in antimicrobial therapy. The TAT approach to the treatment of *S. pneumoniae* infection was studied by evaluating the susceptibility of *S. pneumoniae* to radiolabeled antibody *in vitro* and in an animal infection model. Incubation of *S. pneumoniae* with ^{213}Bi -labelled human mAbD11 resulted in dose-dependent killing of bacteria. TAT of *S. pneumoniae* infection in C57BL/6 mice showed that 60% more mice survived in the ^{213}Bi -D11 treated group than in the untreated group, and hematological toxicity was not observed. This feasibility study has established that TAT can be applied to the treatment of bacterial infections. The treatment of fungal infections by TAT was studied in an animal model of *C. neoformans* infection. Evaluation of acute hematological and long-term pulmonary toxicity showed only transient toxicity in infected mice receiving up to 150 μCi ^{213}Bi -18B7. No evidence of radiation induced fibrosis was found in lungs of treated mice, thus showing the safety of this approach.

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5.3 Radiobiology

The radiobiology programme was started two years ago in an attempt to improve understanding of the cellular and molecular effects of radiation and to support the de-

velopment of procedures for treating patients with radioactive sources. In 2004, studies were continued *in vitro* using human lymphoma (Karpas 422, K1106P) and leukemia (HL60) cell lines, as well as peripheral blood lymphocytes from patients with chronic lymphocytic leukemia (CLL). For tumour cell targeting, the monoclonal antibodies anti-CD20, HuM195 and c595 have been chelated with CHXA''-DTPA and radiolabelled with ^{213}Bi . The responses to alpha-exposure have been examined at different levels of cellular organization: cellular functional level including inhibition of cell proliferation and induction of apoptosis, proteome and transcriptome.

In a common project of ITU, the University of Düsseldorf, Department of Hematology, Oncology and Clinical Immunology and the University of Ghent, we described the specific gene expression profiles induced by alpha irradiation in comparison with gamma rays in human CLL cells by using cDNA arrays and identified 66 genes significantly differentially expressed. These data have been confirmed by real-time reverse-transcriptase-polymerase chain reaction.

In the frame of a project supported by the German Ministry for Education and Research (Bundesministerium für Bildung und Forschung, BMBF), we continued to study, in collaboration with the University of Düsseldorf, the mechanisms of cytotoxicity induced by alpha particles in human normal and malignant hematopoietic (blood-forming) cells. To this aim, we switched from the initially used cDNA Clontech arrays comprising 1,185 genes to the Affymetrix technology. This allowed us a broader screening of the molecular changes contributing to the overall cellular radiation response by examining about 8,500 genes at once. Based on these data, we drew up the first time-dependent and dose-dependent patterns of alpha-induced effects in lymphoma cell lines at transcriptional level.

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4th Alpha-Immunotherapy symposium

The 4th Alpha-Immunotherapy symposium, held in Düsseldorf, June 28-29, 2004, was organised by ITU in co-operation with the Heinrich Heine University of Düsseldorf and the German Cancer Research Center, Heidelberg and is described in the "events" section of this report. The Symposium proceedings are available on CD-ROM upon request.

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Collaborations

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Germany: R. Haas, R. Kronenwett (Universitätsklinikum Düsseldorf); G. Moldenhauer (Deutsches Krebsforschungszentrum, Heidelberg); R. Senekowitsch-Schmidtke (Technische Univ. München)

Italy: K. Abbas, U. Holzwarth (JRC, Institute for Health and Consumer Protection, Ispra)

Switzerland: H. Mäcke (Kantonsspital Basel)

USA: M.W. Brechbiel (National Institute of Health, Bethesda); E. Dadachova (Albert Einstein College of Medicine, New York); D.A. Scheinberg (Memorial Sloan Kettering Cancer Center, New York)

6. Measurement of Radioactivity in the Environment

Scientific and technical activities in support the policy of the Directorate General for Transport and Energy of the European Commission have been continued. These activities concern the implementation of the requirements of environmental radioactivity surveillance (Art. 35-36 of the Euratom Treaty) as well as the framework of the OSPAR (Oslo-Paris Convention) strategy on the management of radioactive substances for the protection of marine environment of the North-East Atlantic.

In the framework of the Enlargement of the EU, the project on "Harmonisation of Techniques and Methodologies for Measuring Radioactivity in the Environment" started in 2003 has been successfully continued. Laboratories from nineteen countries are participating. During 2004 two meetings have been held to discuss the activities of the Network in the project and the results obtained. A training course on "Mass spectrometry (ICPMS) for determination of radioisotopes" has been conducted and twenty-four participants from five different countries attended. The theoretical part took place in Budapest from the 27th to the 29th of April and was jointly organised with the Eötvös Loránd University in Budapest. The practical part was followed in Karlsruhe from the 11th to the 15th of October. From the 8th to the 10th of December an international workshop on "Harmonisation of sampling & measurement techniques for the control of radioactively contaminated soils" was jointly organised with TAIEX (DG ENLARG) in Karlsruhe. About

seventy participants from different nineteen Member States, Accession Countries and Western Balkan Countries attended the lectures given by fifteen invited speakers from different Member States and by DG TREN H4. Two draft reports on "Methods of sampling of soils in cases of radiological alarm or routine verification" and on "Analytical techniques for radiological alarm", respectively, have been finished in 2004 and will be revised during 2005 before final publication as EUR reports.

6.1 Measurements of Radioactivity in Bulk Samples

Studies on **Naturally Occurring Radioactive Material (NORM)** have been performed in view of establishing fast analytical methods for their determination. The initial focus has been on Po-210 and Pb-210, for which radiometric separations have been established. The measurement is performed by alpha spectrometry and by HR-ICP-MS for Po-210 and Pb-210, respectively.

In the framework of support to the Decommissioning and Waste Management Programme, several matrices (soil, sand, wood, bitumen, graphite, lead, steel, etc.) have been studied for the determination of low level of activities of radioisotopes by destructive methods.

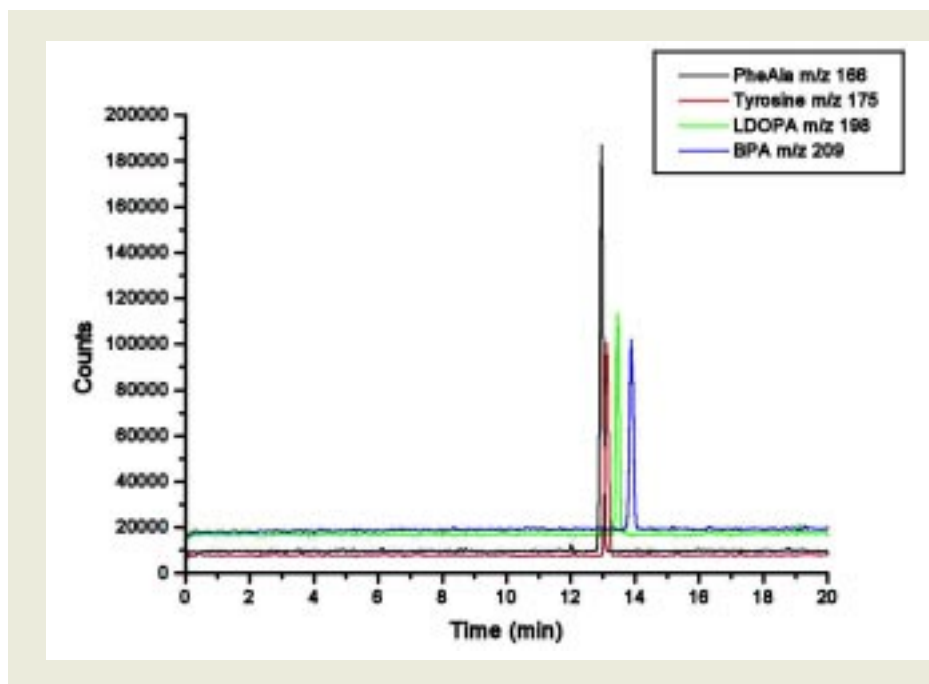


Fig. 1: CE-ESI-MS selective ion electropherograms of phenylalanine, tyrosine, L-DOPA and ¹⁰BPA

6. Measurement of Radioactivity in the Environment

Capillary electrophoresis (CE) conditions have been optimized for the separation of fission products (Lanthanides). Detection was performed by UV spectrophotometry and electrospray mass spectrometry. Capillary electrophoresis separation, compared with the chromatographic separation, presents the advantage as only a microvolume (some nl) of sample is necessary for the measurement. Moreover, structural information on fission products in solution can be obtained by capillary electrophoresis-electrospray mass spectrometry.

In the framework of a collaboration with the Istituto di Fisiologia Clinica (CNR, Pisa, Italy), capillary electrophoresis-electrospray mass spectrometry and HR-ICP-MS have been applied for the detection and quantification of Borophenylalanine (BPA) in biological samples of limited size as applicable for Boron Neutron Capture Therapy. BPA is a modified amino acid, tyrosine analogue. By using CE, ionic species are separated on the basis of their charge and size. In Fig. 1 the separation among phenylalanine, tyrosine, L-DOPA and BPA as obtained by CE-ESI-MS is illustrated. BPA can be rapidly analysed at low concentrations (femtogram).

^{10}B -BPA concentrations are analysed by both CE-ESI-MS and HR-ICP-MS for method validation. Medium resolution has been used for ^{10}B analysis by HR-ICP-MS to avoid interference with $^{40}\text{Ar}^{4+}$ on the mass m/z 10. Good agreement for ^{10}B -BPA determination is obtained between the values from CE-ESI-MS and those from HR-ICPMS.

In Table 1 the results obtained for the determination of ^{10}B -BPA in four samples of cytoplasmatic extracts of cells by CE-ESI-MS and HR-ICP-MS are shown. As it can be seen, they are in a very good agreement.

Sample	CE-ESI-MS (ng/mL ^{10}B)	ICP-MS (ng/mL ^{10}B)
S3	224.7 ± 11.1	224.2 ± 5.6
S7	2189.2 ± 62.0	2190.0 ± 26.3
S12	157.7 ± 4.5	159.9 ± 5.9
S13	497.7 ± 11.9	494.8 ± 14.6

Table 1: Results obtained for ^{10}B -BPA determination in cytoplasmatic extracts of cells by CE-ESI-MS and HR-ICP-MS

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HIGHLIGHT

Characterisation of Environmental U- and Pu-rich Particles by Synchrotron Radiation Based Techniques¹.

In the last ten years, a more thorough examination of nuclear or other radioactive material released into the environment or illegally possessed has become possible.

In the majority of the releases the radioactive material is dispersed into the environment as discrete particles. The relatively few large particles, also called hot particles, carry the majority of the released activity and act like radioactive point sources in the environment. Therefore, it is important to study their environmental impact in terms of mobility, weathering, and corrosion rates. For this, the identification of the source term is necessary. The elemental mapping of the major constituents as well as the content and distribution of trace elements needs to be determined. From the structure of the particle, in terms of distribution of the chemical elements, the origin and the process used for the production of the material can be revealed. Moreover, information on the history of the material in terms of preferential leachability and, therefore, transfer to the food chain can be obtained. In relation to this, the elemental speciation plays one of the most important roles. Chemical speciation is critical towards understanding and predicting the fate and transport in the environment of radionuclides as well as their bio-availability. For this scope, the elemental mapping distribution of the radionuclides, their oxidation states (in the case of U, Pu, Np and Am) as well as their 3D distribution need to be studied.

Environmental U- and Pu- rich particles have been extensively studied following the analytical approach as sketched in Fig. 1 HL. After their separation from bulk materials, they have been located with the appropriate

¹ These investigations were performed in collaboration with the KFKI-Atomic Research Institute (Hungary), IAEA- Agency's Laboratories Seibersdorf (Austria), Risø National Laboratory (Denmark), Atominstutut der Österreichischen Universitäten, Technische Universitäten Wien (Austria), Synchrotron facility at HASYLAB and at ANKA.

detection techniques and then characterised, for their elemental composition, U/Pu mapping and 3D distribution.

Before analysing the particles by microscopic X-ray fluorescence (μ -XRF) at the FLUO-TOPO beam line at the ANKA synchrotron facility (FZK, Karlsruhe), they have been examined by scanning electron microscopy (SEM) combined with energy-dispersive X-ray (EDX) or wavelength-dispersive X-ray (WDX) spectrometry. This is of particular interest when the particles are supposed to contain a mixture of U and Pu, revealing the surface and to some extent subsurface composition. Utilising μ -XRF at a synchrotron radiation (SR) facility, information on the elemental composition of the particles has been obtained. When applying a 20 μ m beam, the distri-

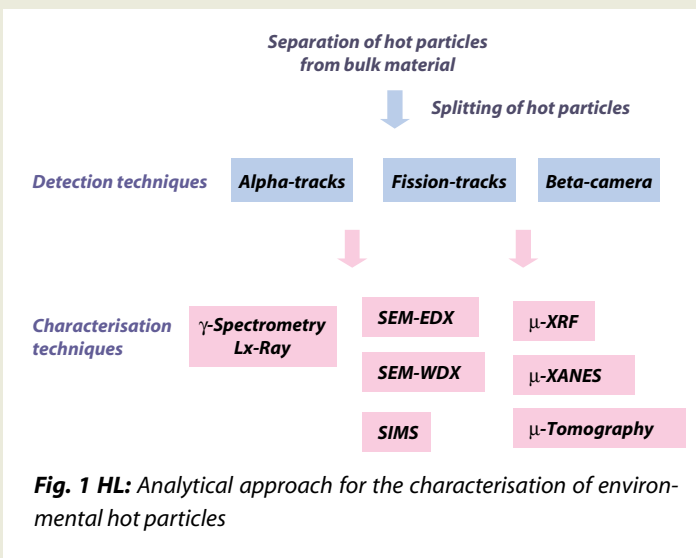


Fig. 1 HL: Analytical approach for the characterisation of environmental hot particles

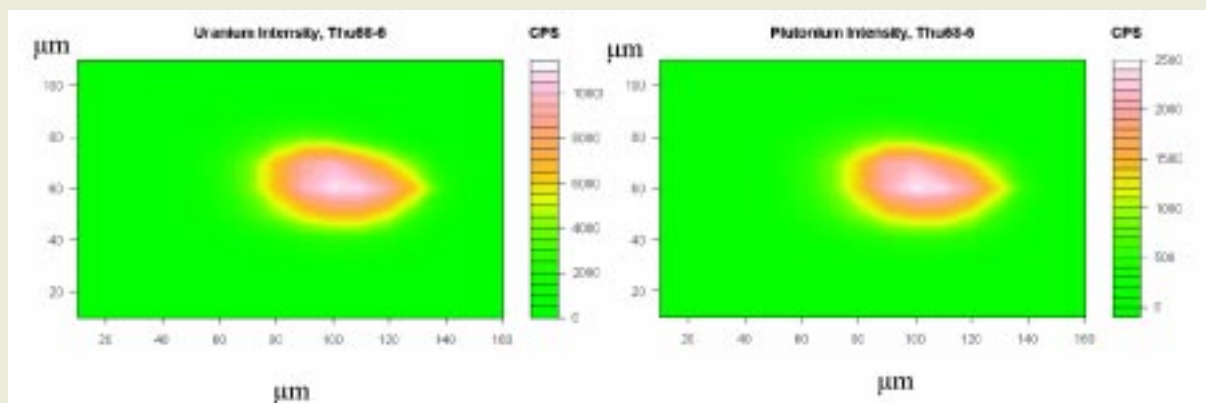


Fig. 2 HL: Intensity distribution of Pu and U in a single particle by μ -XRF at FLUO-TOPO beam line in ANKA (beam diameter 20 μ m)

bution of U and Pu in the particles is revealed as shown in Fig. 2 HL.

Of special interest were the results obtained by using a beam size of 2.5 μ m. In these experimental conditions micro-inclusions in a large object can be observed and studies on the heterogeneity of the material can be carried. Detailed information on the microstructure can then provide information on the production process of the original material. In Fig. 3 HL a large particle is shown with the zone selected for detailed investigation. The results obtained for the fine distribution of U and Pu in the investigated area are shown as an example in Fig. 4 HL.

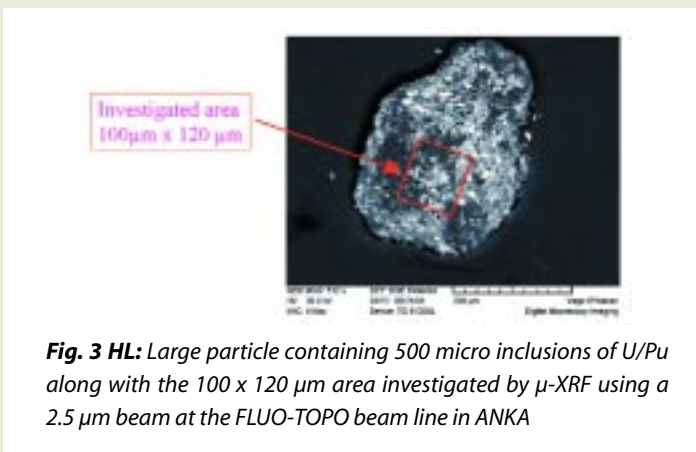


Fig. 3 HL: Large particle containing 500 micro inclusions of U/Pu along with the 100 x 120 μ m area investigated by μ -XRF using a 2.5 μ m beam at the FLUO-TOPO beam line in ANKA

6. Measurement of Radioactivity in the Environment

Microscopic X-ray absorption near-edge structure (μ -XANES) has been exploited to study the oxidation states of uranium and plutonium in U/Pu mixed particles. The experiments were performed at the beam line L at HASYLAB. Five different particles containing U and Pu along with the standards samples were measured. The distribution of U and Pu between oxidation state IV and VI is summarised in Table 1 HL.

Particle ID	U(IV)%	U(VI)%	Pu(IV)%	Pu(VI)%
3	90.9	9.1	92.0	8.0
5	90.9	9.1	97.0	3.0
6	100.0	0	25.0	75.0
4-5	75.0	25.0	33.0	67.0

Table 1 HL: Summary of the results obtained from μ -XANES measurements for the oxidation states of U and Pu

Combined XRF and absorption μ -tomography was applied at the FLUO-TOPO beam line in ANKA to obtain the 3D U/Pu chemical distribution in the particles. In Fig. 5 HL the reconstructed 3D distribution of U and Pu in a particle recovered from sea sediment. The plutonium (blue) and uranium (green) are coated with iron (red) rich sediment matrix.

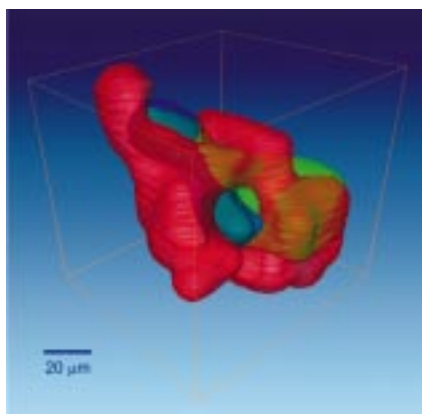


Fig. 5 HL: Reconstructed 3D distribution of elements in U/Pu-rich particle recovered from a sea sediment. The plutonium (blue) and uranium (green) are coated with iron (red) rich sediment matrix.

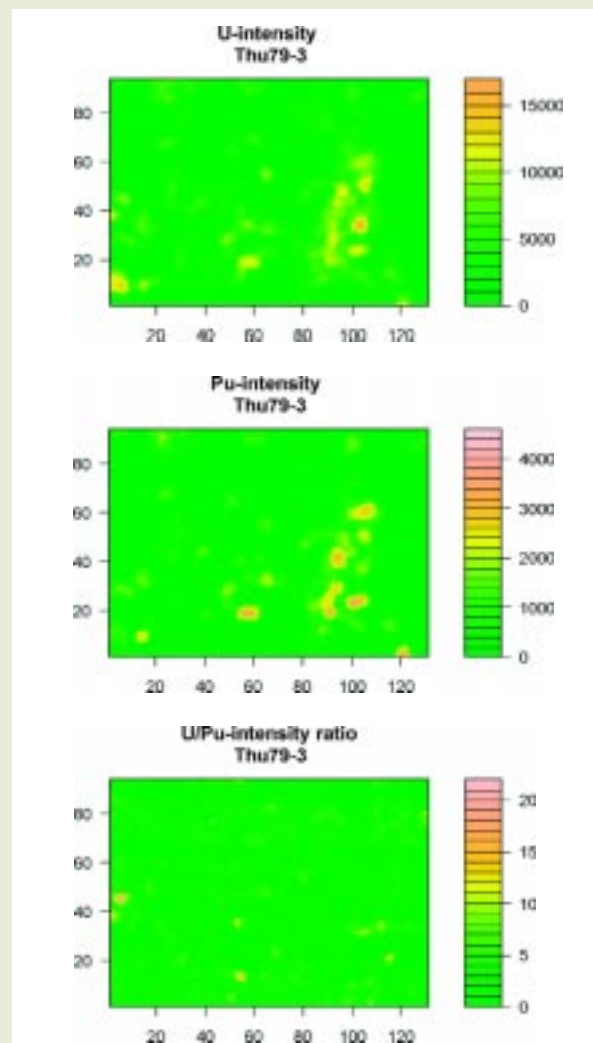


Fig. 4 HL: μ -XRF mapping of an area of $100 \times 120 \mu\text{m}$ by using a $2.5 \mu\text{m}$ beam at the FLUO-TOPO beam line in ANKA. a) U intensity, b) Pu intensity; c) U/Pu intensity ratio.

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6.2 Environmental Radioactive Microparticles

In the field of radioactive environmental micro-particles, methods for dividing samples and isolating the particles have been extensively applied on real samples.

In marine sediments uranium particles have been detected by fission tracks, after irradiation in a TRIGA reactor (see Fig. 2a) and by alpha tracks after different exposure time (see Fig. 2b).

As for the Pu containing particles, the splitting of the samples was performed following with a gamma detector, the signal due to Am-241 and the particles containing Pu were selected with the use of a beta-camera detector. After that the particles were picked with the help of a micromanipulator.

Once the particles have been isolated, micro X-ray fluorescence analysis was performed at the FLUO-TOPO beam line at the ANKA synchrotron (Karlsruhe, Germany) for the determination of the elemental distribution of Th, U and Pu in the particles.

The same particle was then examined at the beam line L at the DESY HASYLAB in Hamburg as for the determination states of uranium. Uranium was found present in the particles mostly in the U(IV) form.

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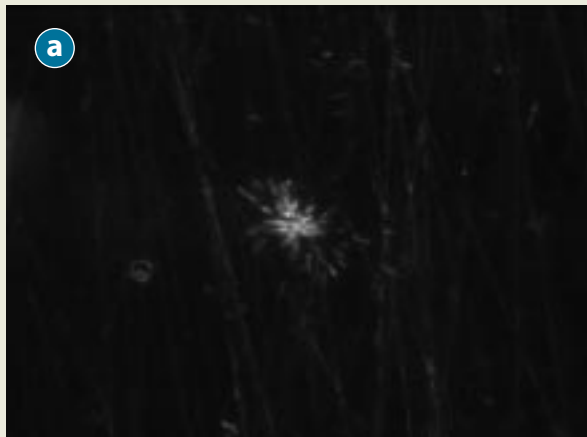
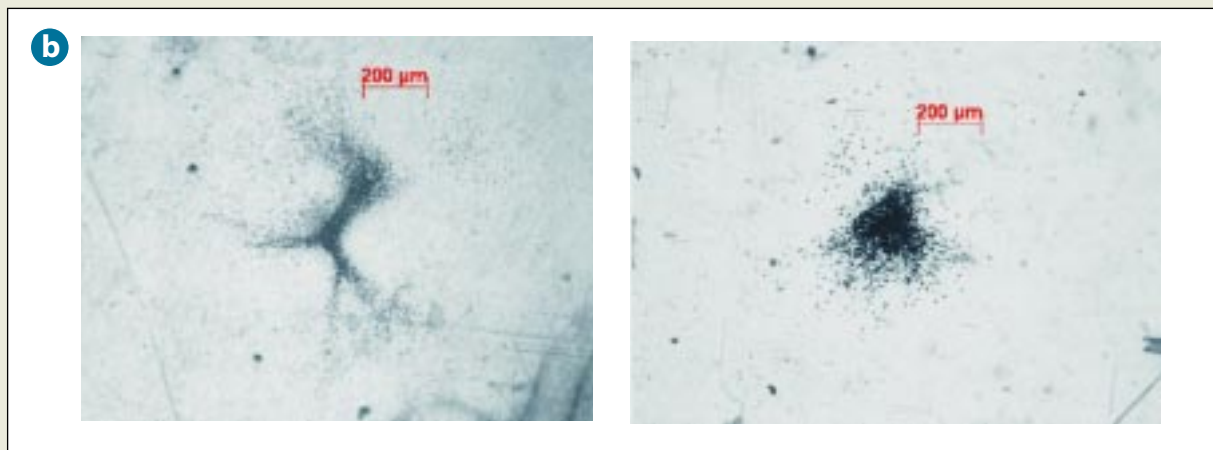


Fig. 2: (a) Fission tracks obtained after irradiation in a TRIGA reactor from a particle containing uranium stemming from marine sediments. (b) Alpha track images for two hot particles stemming from marine sediments. Exposure time 43 days (left side); 8 days (right side).



6. Measurement of Radioactivity in the Environment

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7. Safeguards Research and Development

The development and improvement of measurement methods and their application to samples taken for nuclear safeguards purposes remains the core activity within the nuclear safeguards action. The nature of samples may range from nuclear fuel to microparticles collected in the environment. The development activities and the service measurements related to the latter are addressed in the "Highlight". The activities reflect the specific needs of the main customers (i.e. Euratom Safeguards – DG TREN; International Atomic Energy Agency – IAEA) and take into account the evolution of the nuclear fuel cycle as well as public concerns arising from nuclear terrorist threat. ITU's development activities are embedded wherever necessary in international collaborations, aiming at profiting from synergies and ultimately at further strengthening the Institute's leading position in this area.

7.1 Improvement of Measurement Techniques and Methods

Compucea 2nd generation

A second generation of equipment for the combined uranium concentration and enrichment assay (COMPUCEA) is under development. The goal is to create a truly portable and ready-to-use instrumentation for simplified in-field operation. With the new design it is expected to save time and costs for the in-field missions of ITU analysts, who provide regular measurement support with Compucea to joint DG-TREN/IAEA inspections in LEU (Low enriched uranium) fuel fabrication plants. The development of the 2nd generation Compucea constitutes a task in the support programme to the IAEA.

A prototype of the new Compucea for uranium concentration measurements has been completed for test measurements and performance evaluation. The adopted measurement technique is based on L-edge absorptiometry. The total weight of the equipment (see

Fig. 1) could be significantly reduced to about 6 kg through the use of advanced technology. Major practical benefits accrue from the elimination of detector cooling with liquid nitrogen for simplified in-field operation, and from the substitution of a radioactive source by a low power X-ray unit for easier instrument transportation.

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Calorimetry

Research work on the use of calorimetry in Safeguards for quantitative plutonium assay in small samples has continued. Principal studies further addressed the question of uniqueness of sample heat production, irrespective of given physical sample properties. Previously observed inconsistencies in the specific heat production of sintered and non-sintered plutonium materials could not be confirmed and verified from measurements performed on further materials.

A new small sample calorimeter has been procured and made operational for routine use. The system (model TAM III from Thermometric) presently holds two independent small sample (20 ml) calorimeter units, but can be expanded up to 4 independent units for increased sample throughput. Initial performance studies showed

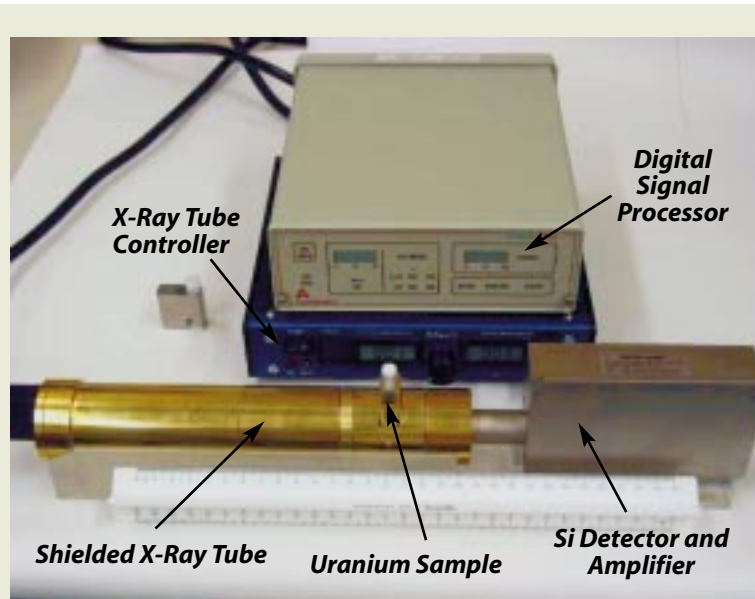


Fig. 1: View of the new Compucea for uranium concentration measurements

continued on p. 64

7. Safeguards Research and Development

HIGHLIGHT

Characterisation of Radioactive Microparticles for Nuclear Safeguards Verifications and Detection of Undeclared Nuclear Activities

After the Gulf war in 1990 and the disclosure of a secret nuclear program in Iraq it became necessary to strengthen the control of nuclear activities by the International Atomic Energy Agency (IAEA) and improve the capability of the early detection of undeclared nuclear activities. In order to evaluate the different options, IAEA launched the program 93+2. One key component of this program, as well as of the HPTA-program (High Performance Trace Analysis) of EURATOM, is the development of analytical techniques to determine the isotopic composition of uranium particles collected in swipe samples. Nowadays, with the implementation of the Additional Protocol swipe samples are taken by IAEA and EURATOM inspectors inside nuclear installations (such as enrichment and reprocessing plants) during routine verifications of nuclear safeguards. The composition of these particles can be used to detect activities, declared or undeclared, in the controlled plants.

Since the beginning of the Program 93+2, as well as the HPTA project, ITU has been collaborating with both IAEA and EURATOM in developing the analytical methodology for the detection of the isotopic compositions of microparticles, as well as in providing results in routine nuclear safeguards verifications. Since 1997 ITU belongs to the Network of Analytical Laboratories of the IAEA.

Secondary Ion Mass Spectrometry (SIMS) was implemented at ITU in 1996 and has been routinely used for the determination of the uranium isotopic composition in microparticles collected in swipe samples. During 2004 the number of such samples drastically increased, from 50 such in 2003 to 160. To assure continued high-quality results

for the customers (IAEA, DG TREN, ABACC and KAERI) an internal Quality Control Programme is maintained. This programme is based on the periodic analysis of two types of quality control samples. Blank swipe samples are analysed to detect possible contamination and cross-contamination during the analysis process, while well characterised standard uranium particles are measured to check the level of statistical control.

A validation study using statistical data analysis on measurement results obtained with the SIMS CAMECA 4F on a set of Certified Reference Materials (CRM) from NIST, characterised by homogeneous 1 µm particles with different ^{235}U enrichments (1, 2, and 10 %) has been performed. Using these data it is also possible to estimate the measurement uncertainty according to the GUM (ISO Guide for the Expression of Uncertainty in Measurement). The evaluation of the results obtained is an excellent indicator for the quality of the SIMS for uranium isotopic analysis. The proven reliability of the different parameters monitored was recently successfully used for the accreditation of the method according to ISO 17025. In Fig. 1 HL a quality control chart for a 10% enriched reference material is shown.

Studies on particle extraction procedures from swipes as well as implementation of the performance of the SIMS instrument and Quality Control measures have been performed. In particular, in order to locate interesting particles

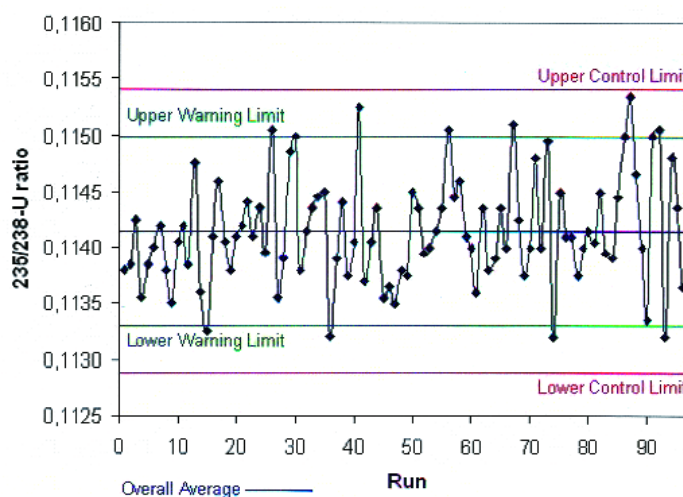


Fig. 1 HL: Quality Control Chart: measured $^{235}\text{U}/^{238}\text{U}$ ratio for particles of a certified reference material

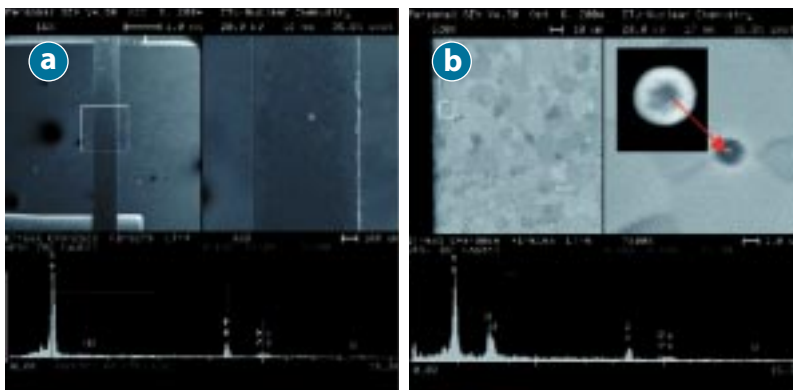


Fig. 2a+b HL: These pictures from scanning electron microscopy (SEM) show a uranium reference particle after micromanipulation and deposition on a TIMS Rhenium filament. The particles were analyzed by SEM (upper part) with different magnifications increasing from the left to the right. The lower part of the pictures shows the X-ray fluorescence spectrum, with the characteristic peaks of uranium and rhenium.

in samples with low density of actinides and determine their isotopic composition, complementary techniques, based on the fission track method and thermal ionisation mass spectrometry (FT-TIMS), are being developed.

A procedure for rapid recognition of ^{235}U -enriched particles using this technique was developed as a first step in an analytical procedure for the survey of environmental particles. The main purpose is to use the solid state nuclear track detection (SSNTD) method to locate interesting particles,

as well as a first estimation of their activity and content of fissile material. For U-containing particles of a given size, there is a correlation between the number of tracks and the enrichment level. Among the different SSNTD tested, Lexan and Makrofol showed the possibility of distinguishing 3, 10 and 50% ^{235}U enriched particles after only 1 minute of irradiation. In order to obtain qualitative distinction of particles from 1 to 50% enrichment 3 minutes of irradiation were sufficient.

After irradiation, the particles need to be correlated with the tracks developed in the SSNTD and prepared for the next measurement. In particular, a methodology to micromanipulate the particles and then to deposit them on a filament for analysis by TIMS to measure their isotopic composition has been developed. In Fig. 2 HL a uranium reference particle is analysed by SEM after micromanipulation and deposition on a TIMS Rhenium filament.

Techniques for manipulating micrometer-sized particles in a scanning electron microscope are also important for research related to safeguards and forensic purposes. Uranium oxide microparticles ranging from sub- μm to several 10 μm can be freely manipulated by adhering them to the tip of a probe. In Fig. 3 HL the micromanipulator system used for precision positioning and collecting individual particles by means of using microtips (down to 100 nm) is shown.

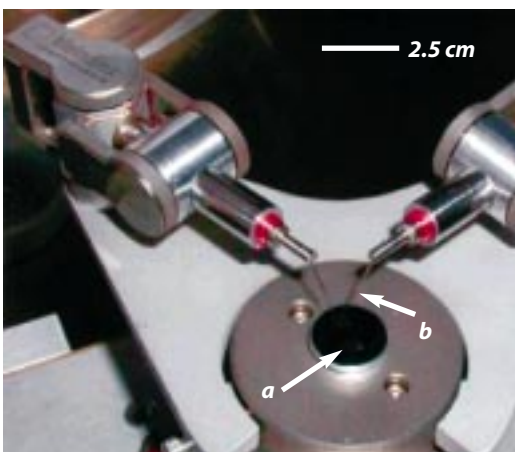


Fig. 3 HL: Micromanipulator system installed under ultra high vacuum on a scanning electron microscope. It has 2 rotational, 1 linear axes and it is based on piezomotor technology with sub nanometer precision. The arrow "a" shows the carbon planchet where the particles are deposited, while the arrow "b" shows the microtips operating at the surface of the sample.

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7. Safeguards Research and Development

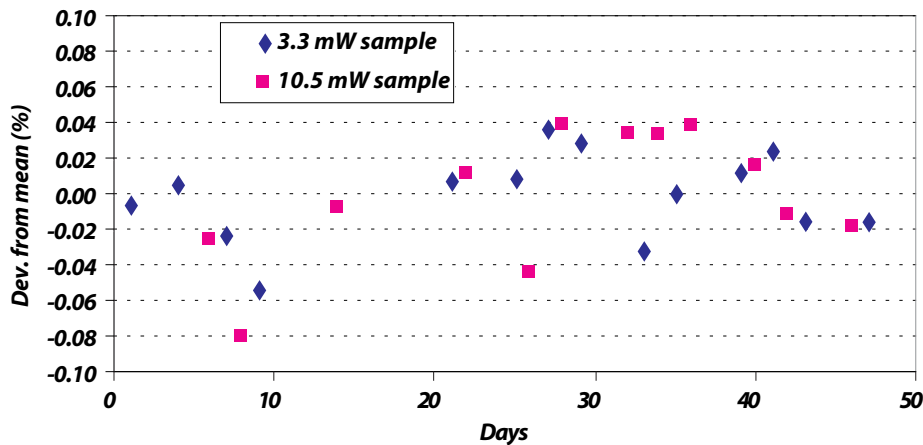


Fig. 2:
Measurement reproducibility observed with the new small sample calorimeter

excellent measurement reproducibility with standard deviations of better than 0.05 % for sample powers in the mW range (see Fig. 2).

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Analytical control of the actinides in the pyroprocessing

Pyrometallurgy is a promising alternative to aqueous partitioning of spent nuclear fuel. Process development and quality control of pyrometallurgy require analysis of the separated salt and metal phases. In addition, the experience gained from these analyses is of key importance for establishing effective safeguards of a future P&T fuel cycle. At ITU, radiometric assay techniques such as X-ray fluorescence (XRF), neutron coincidence counting (NCC) and high-resolution gamma spectrometry (HRGS) are employed for the quantitative determination of actinide elements (U, Np, Pu, Am, Cm) in the process samples. An alternative technique for elemental analysis is inductively coupled plasma mass spectrometry (ICP-MS). In particular the heavier elements can be detected with very good sensitivity. Comparative measurements of 12 samples from both metal and salt phases were carried out and the amount of U, Pu, Am and Cm was determined (Table 1). The results from both methods are in good agreement within statistical uncertainties which validates the analytical approach to this new separation technique.

Design work for a new NDA station combining the radiometric techniques of XRF, NCC and HRGS for the direct actinide/lanthanide assay in sample specimens from the pyroprocessing of irradiated fuels has been started. The new NDA measurement station will be directly attached to the hot cell facility of ITU.

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7.2 Detection Methods for Measurement of Microparticles

In order to reduce the measurement uncertainty and to improve the accuracy of the particle techniques, standard particles with a well known content of uranium and plutonium are needed. These particles can allow the detection efficiency to be determined. Furthermore, reference particles with a well-defined isotopic composition are required for the quality control by mass spectrometry measurement. ITU has successfully produced monodisperse uranium oxide particles starting from reference materials of different uranium isotopic composition using a commercial vibrating orifice aerosol generator [1].

Sample	U			Pu			Am			Cm		
	ID	ICPMS	XRF	% diff.	ICPMS	XRF	% diff.	ICPMS	HRGS	% diff.	ICPMS	NCC
B49	1063	721	-32	1365	1244	-8.9	148	146	-1.1	34.7	36.5	5.3
B45	2635	2305	-13	2297	2221	-3.3	255	244	-4.2	50.0	51.8	3.6
B61	1071	633	-41	1140	881	-23	136	135	-0.8	29.0	30.9	6.4
B63	218	< det lim	-	142	< det lim	-	16.2	16.6	2.7	2.36	2.40	1.7
B65	1073	881	-18	850	699	-18	90.0	93.4	3.8	16.3	17.1	4.8
B60	278	< det lim	-	98.5	< det lim	-	10.1	10.3	2.4	2.50	2.75	10
S50	7161	6979	-2.5	3345	3388	1.3	380	342	-10	90.2	80.2	-11
S57	3249	2675	-18	1500	1453	-3.1	189	174	-8.1	65.8	64.4	-2.1
S63	1448	1340	-7.4	1617	1597	-1.2	252	218	-13	91.7	86.6	-5.5
B52	3627	3459	-4.6	1297	1220	-5.9	128	119	-7.4	22.6	24.0	6.3
B64	3660	3357	-8.3	580	519	-11	42.0	32.4	-23	8.14	7.23	-11
B67	1386	1256	-9.4	852	795	-6.6	98.0	88.7	-9.5	20.1	21.5	6.8

Table 1: Absolute concentrations of the actinide elements (in mg/g) and the relative differences of results obtained for the same sub-samples measured by different analytical techniques. Cm was measured by neutron coincidence counting, Am by gamma spectrometry, U and Pu by XRF.

During 2004 a new campaign of production of 1 µm size monodisperse uranium oxide particles starting from 3% enriched NBL (New Brunswick Laboratory) uranium reference material was carried out. Since the same system will be exploited for the production of standard particles containing plutonium and uranium/plutonium mixtures, after these tests the apparatus was dismantled and completely refurbished to be inserted in a new glove box.

Another parameter relevant to nuclear safeguards or the nuclear forensics is the age of U or Pu particles. Age determination of Pu particles by SIMS has been demonstrated earlier [2]. The age of such a particle can be determined on the basis of the parent/daughter ratios $^{231}\text{Pa}/^{235}\text{U}$ and $^{230}\text{Th}/^{234}\text{U}$ established by radioactive decay of the respective parent nuclide [3]. In this work, the mass spectrometric signals had to be corrected for the background contribution and the assumption that no difference in the ionisation efficiency of Th, Pa and U was made. During 2004, standard uranium oxide materials with known ages were used to check for a possible difference in the ionisation efficiency of the 3 elements. The first results obtained on NBL uranium oxide standards

seem to confirm the results obtained earlier. No sensitive difference in the ionisation efficiency of Th, Pa and U was seen. Better results are obtained with the ratio $^{230}\text{Th}/^{234}\text{U}$ because it does not need any background correction and only one detector is used to register the intensity signals of the 2 isotopes.

References

- [1] N. Erdmann et al., Spectrom. Chim. Acta B 55(2000) 1565
- [2] M. Wallenius et al., Radiochim. Acta 89 (2001) 55
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7. Safeguards Research and Development

7.3 Analytical Support

The accreditation of the Analytical Services according to the standards set by ISO 17025 is described earlier in this report.

ITU implemented Quality Management according to ISO 9001 already in the mid 1990's. Much more stringent requirements, targeted at testing laboratories, are imposed by ISO 17025. The Analytical Services (AS) section has achieved accreditation according to this standard. Accreditation is a third-party attestation that conveys formal demonstration of the competence to carry out specific analyses. Following an audit in November 2004, the DACH (Deutsche Akkreditierungsstelle Chemie GmbH) has granted accreditation to carry out testing in the field of chemistry using radiometric methods, titration, and mass spectrometry for the testing of U and Pu bearing materials, aqueous and acidic solutions and swipe samples.

Following the recommendation of the High Level Experts Group the principle of High Performance Trace Analysis (HPTA) is not only maintained for the determination of the uranium isotopic balance as the basis for accounting verification in an enrichment facility but is also used for the detection of undeclared HEU production activities. The isotopic composition of uranium particles from swipe samples collected inside enrichment plants is measured using Secondary Ion Mass Spectrometry (SIMS). In the reporting period the isotopic compositions of uranium particles in 116 swipe samples (originating from IAEA, DG TREN, ABACC and KAERI) were determined. The increase of this activity over the last three years is shown in Fig. 3.

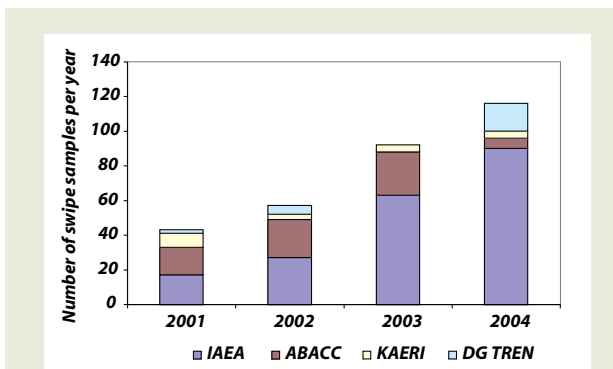


Fig. 3: Evolution of number of swipe samples analysed over the last three years, broken down per customer

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In-field analysis

64 uranium samples (pellets, powders and scraps) were measured in different European uranium fuel fabrication plants (Sweden, Belgium, Spain and France) using the mobile Compucea instruments. These measurement campaigns are in direct support to DG TREN, when carrying out the Physical Inventory Verification in the plants. The results are normally available within three to four days. Table 2 indicates the high performance achieved in the campaigns of 2004, which is in full accordance with the International Target Values for this kind of measurement.

Year 2004	U content		²³⁵ U/U	
	Pellet	Powder	Pellet	Powder
Bias%	0.01	-0.02	-0.04	-0.03
Stdev	0.12	0.27	0.40	0.55
Target Value	0.25	0.25	0.45	0.45

Table 2: Compucea performance 2004; Average difference (bias) between declared values and Compucea measurement results, variability (Stdev) and the IAEA International Target Values are shown for comparison

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7.4 Illicit Trafficking and Nuclear Forensic Science

Nal gamma spectrometry of shielded uranium/ limits and new approach of uranium identification

The detection of uranium is of greatest interest because it is the type of nuclear material mostly encountered in illicit trafficking. However, its detection and identification are generally difficult and sometimes nearly impossible, especially if the uranium is highly enriched and concealed under shielding. Comprehensive studies were performed on the appearance of uranium gamma spectra seen by a typical hand-held monitor equipped with a NaI detector through various shielding materials. Result-

ing gamma spectra were both obtained experimentally and by Monte Carlo simulation studies (see Fig. 4). From a review of the spectral data we propose a new approach for the identification of uranium which makes also use of the shape of the measured gamma continuum observed from shielded uranium. This complementary information can be helpful in cases of poor statistical data for the full photon energy peaks of uranium gamma rays. The applicability and limitations of this analysis approach is still under study.

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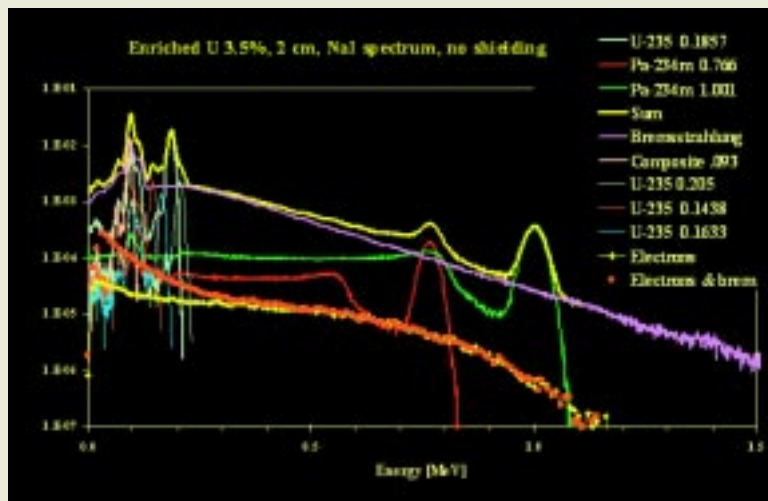


Fig. 4: Contribution of uranium isotopes and decay daughters to the observed uranium gamma spectra

Examination of the Dounreay Fast Reactor Particles

In a follow up to the investigation of the Dounreay Materials Test Reactor (MTR) particles, which has now been completed, five particles of a different composition are being examined in the Institute. These particles were recovered from the sea bed and foreshore, and originate from the Dounreay Fast Reactor (DFR) which used a uranium-molybdenum alloy fuel. The particles, however, consist of U, Nb, and Fe with small amounts of Al, Cs and Si, as analysed by Energy Dispersive X-ray analysis (EDX) in the Scanning Electron Microscope (SEM). Mo was absent, it could not be detected in the EDX analysis.

The particles are smaller than the MTR particles, ranging from 0.1 mm to 0.8 mm along the major axis, and are friable. The number of particles recovered is very much lower than that of the MTR material.

Figure 5 shows a back-scattered electron image of particle LSN 970146 recorded in the Philips XL40 SEM, showing it to be fragmented and highly porous. The EDX analysis performed on this particle confirmed the presence of U, Nb and Fe, but the absence of Mo, which was a major constituent of the original fuel.

The particles probably result from an accident which is known to have occurred at Dounreay during the dissolution of the fuel as a first step to reprocessing. Segments of the fuel were to be dissolved in a Nb dissolver vessel when the reaction went out of control, leading to a high

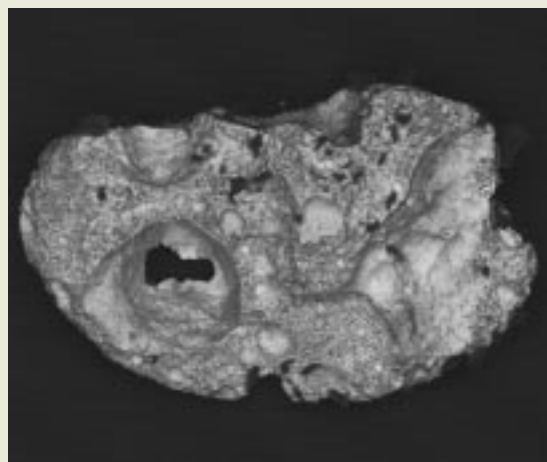


Fig. 5: Backscattered Electron Image of Dounreay DFR particle LSN 970146 recorded in the Philips XL40 Scanning Electron Microscope

temperature increase and an explosive rupture of the vessel. The highly porous nature of the particles indicates that the Mo was lost from the fuel at this stage by evaporation as MoO_3 . The investigations are continuing on this and the other four particles, and will be complemented by EPMA and SIMS analyses.

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7. Safeguards Research and Development

Supporting new member states in combating illicit trafficking of nuclear materials

In the framework of the EU enlargement and integration activities, ITU continued its cooperation with the new EU member states on combating illicit trafficking. Demonstration exercises were organised in Poland and in Slovakia, in order to verify the proper implementation of the national response plan. A workshop reviewing the "Experience in combating illicit trafficking" was held in Vilnius with participants from the new EU member states, from western Balkan countries and from the International Technical Working Group on nuclear smuggling, the IAEA and Europol.

Co-operation with Russia

The co-operation with the Bochvar Institute (VNIINM) in Moscow was continued in the framework of the Commission's TACIS programme. Data processing equipment was delivered to VNIINM and an inductively coupled plasma mass spectrometer (ICP-MS) dedicated for analysis of impurities in uranium plutonium was tested at the factory and is being delivered to Moscow. An ICP-MS hands-on workshop was held for analysts from VNIINM.

The existing database system for support of analyses of nuclear material of unknown origin (TUAR-96, p.135 and TUAR-98, p.33-35, 104) was maintained. A new agreement has been signed between a large European supplier of nuclear fuel and ITU Karlsruhe. It assures the transfer of characteristic data for UO₂ fuel pellets manufactured up to 2009, once per calendar year. The data cover:

- fuel pellet geometry
- initial U isotope concentrations
- concentrations of characteristic impurities
- distributions of grain and pore sizes

Continuing the long-term co-operation with the All-Russia Institute for Inorganic Materials (Bochvar-Institute/VNIINM) Moscow, two installations were maintained:

- the common sector of the relational database at VNIINM Moscow, symmetric to that installed at ITU
- the electronic literature archive at ITU Karlsruhe (TUAR-2003, p. 69). This TACIS-Bistro project was completed in the reporting period.

In the frame of an ongoing TACIS contract (AA 2002/030-307), a new subcontract between VNIINM and ITU has been signed. It assures a regular update of characteristic data for Russian nuclear fuel up to 2006.

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Radiological Dispersal Devices

The international community is concerned about the possible criminal use of radioactive and nuclear substances (UN resolution 1540), which in a Radiological Dispersal Device (RDD), also known as "Dirty Bomb". ITU is cooperating with IPSC and the German Federal Office for Radiation Protection (BfS) to predict the effects of such an attack at any site within Europe. The modelling code "LASAIR", which describes the inhalation dose and the afflicted area after a Radiological Dispersal Event (RDE), could be significantly improved enabling the use of digital satellite images. Hence, it is applicable to every location of known topography. The results of this work were presented at several occasions and have been appreciated by the United Nations Interregional Crime and Justice Research Institute (UNICRI).

Reference

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7.5 Operation of the Laboratoria In Situ

In order to assure timely, efficient and effective safeguards verification measurements for the two large reprocessing plants in Europe, Euratom Safeguards (DG TREN) installed analytical laboratories at the respective sites. Both laboratories were designed and commissioned by ITU and have been successfully operated now for several years by specialised ITU staff.

Laboratoire Sur Site (LSS), La Hague

The EURATOM laboratory located on the site of the French reprocessing plant at Cap de La Hague and inaugurated on the 6th of June 2000 (TUAR-00, p. 24-25) was designed to perform analysis of input and product samples in the framework of EURATOM safeguards for nuclear material verification (TUAR-03, p. 70-71).

During this fourth full year of operation, the laboratory continued to be manned by experienced ITU analysts who carried out a total of 182 mission weeks on site (i.e. an average of 3 analysts/week) to perform analytical work, maintenance, administrative tasks and to follow mandatory (re-) training courses.

During the 48 calendar weeks of presence of ITU analysts, 798 samples have been analysed representing an increase of 22% as compared to the previous year. A detailed list is given in Tables 3 and 4, respectively.

Sample type	Measured samples
Input	413
UCD	112
URP (« old Pu »)	134
PuO ₂	100
Inventory samples	36
“Occasional” samples	3
TOTAL	798

Tab. 3

Measurement type	Measurements
HKED	1643
density	805
Chemical separations	333
Mass spectrometer	844
Gamma	104

Tab. 4

The Hybrid K-edge instruments and gamma detectors are the *workhorses* of the LSS. These radiometric methods are commonly named Non Destructive Analysis (NDA) and they have performed the measurements of all samples transferred to the laboratory.

Due to logistical problems, beyond the control of ITU, transport of nuclear reference material could not be organized by DG TREN in 2004. Consequently, ITU needed to economise on the consumption of spikes by reducing the number of QC measurements by Isotope Dilution Mass Spectrometry (IDMS). IDMS was only applied to samples where discrepancies between LSS NDA results and Operator's results were noticed by Luxembourg Inspectors. Furthermore, IDMS was applied to characterize solutions serving for NDA instrument calibration and to accurately measure nuclear material in specific samples that cannot be analysed straightforward by NDA.

During 2004, tests were carried out to improve the preparation of sources for alpha spectrometry and to study the stability of U and/or Pu solutions used to calibrate the hybrid K-edge instruments.

The recent experience has shown that the workload in the LSS is not simply a function of the number of samples but increasingly depends on the sample type (i.e. there is an increasing amount of “exotic” samples, not initially foreseen, which require much more analytical effort).

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On-Site Laboratory (OSL), Sellafield

The OSL is located at the site of the BNFL reprocessing plant in Sellafield (UK) and performs verification measurements on samples taken by EURATOM inspectors on the site and in UK fuel fabrication plants. The OSL is operated by ITU staff members who performed a total of 134 mission weeks during the year 2004. A total of 208 samples were analysed. Figure 6 shows the number of samples analysed from 2000 till 2004 and the distribution according to the sample type. Radiometric techniques are normally used as “work horses”; i.e. the majority of the samples are measured using these methods, while a subset of samples (10 – 15 %) is measured additionally by mass spectrometry, serving as a reference method.

Due to breakdowns of the radiometric equipment, mostly related to the age of the installation (e.g. replacement of the X-ray tube, and decayed ¹⁰⁹Cd sources), there was a shift in workload from radiometric techniques to Isotope Dilution Mass Spectrometry and Thermal Ionisation Mass Spectrometry. During those periods, all samples have been analysed using mass spectrometry, allowing the OSL to fulfil its commitment. It is apparent that only

7. Safeguards Research and Development

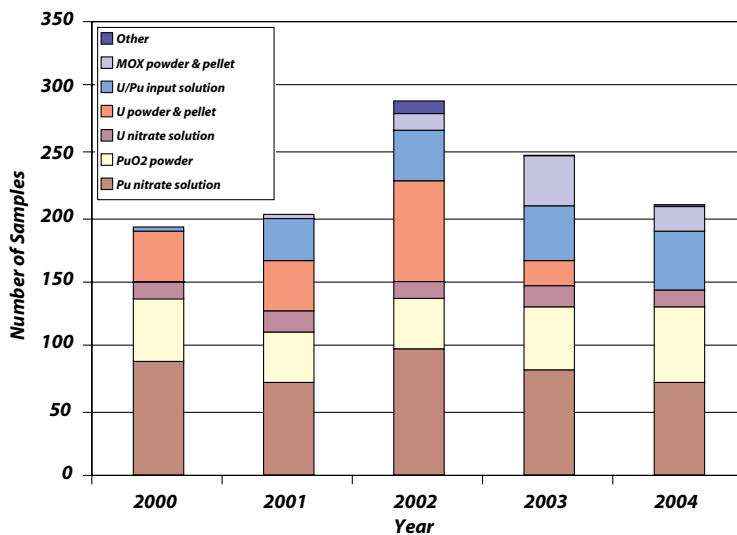


Fig. 6: Number of samples analysed in the OS from 2000 till 2004 and distribution according to sample type

the regular use of mass spectrometry methods under statistical control has allowed this flexibility.

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7.6 Support to the IAEA

Support to the installation of the IAEA on-site laboratory at Rokkasho-mura

The neutron coincidence counter designed and built by ITU on request of the IAEA for the IAEA on-site laboratory at Rokkasho has been completed for final testing and calibration. Figure 7 shows the neutron counter on top of a Hybrid K-Edge Densitometer (HKED) installation at ITU, which exactly reproduces the measurement configuration for the destination of the neutron counter at Rokkasho. For pre-calibration at ITU, ^{244}Cm and ^{252}Cf reference solutions have been prepared and carefully characterised by means of Isotope Dilution Alpha Spectrometry.

On invitation of the IAEA, two measurement specialists of the ITU OS team paid a visit to the on-site laboratory at Rokkasho for a critical review of the adopted analytical procedures. The proposals for modifications have been forwarded to the two organisations that will jointly operate the laboratory, namely IAEA and to NMCC (the Japan-

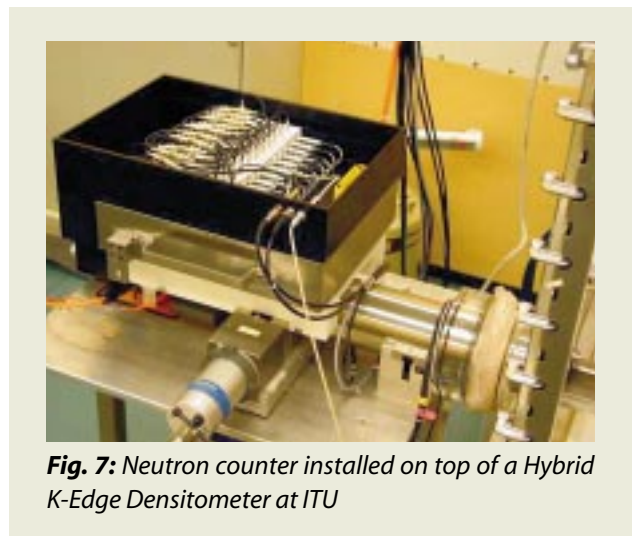


Fig. 7: Neutron counter installed on top of a Hybrid K-Edge Densitometer at ITU

ese Nuclear Material Control Centre). Two IAEA staff members followed training at ITU in the use of the hybrid k-edge instrument. NMCC spent a technical visit to the LSS, La Hague, in order to get a more thorough insight in the operational aspects of the on-site laboratory.

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Overview

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Actions

Basic Actinide Research (F. Wastin/Action Leader)

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Spent Fuel Characterisation (V. Rondinella/Action Leader)

M. Amme, P. Carbol, J. Cobos-Sabate, V. Rondinella, D. Serrano, D. Wegen, T. Wiss

Safety of Nuclear Fuels (C. Walker/Action Leader)

D. Bottomley, S. Brémier, M. Sheindlin, J. Somers, J. Spino, E. Toscano, P. van Uffelen

Partitioning and Transmutation (R. Konings/Action Leader)

B. Christiansen, A. Fernandez, J. Galy, R. Konings, J. Magill, R. Malmbeck, T. Zagar

Alpha-Immunotherapy/Radiobiology (C. Apostolidis/Action Leader)

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Measurement of Radioactivity in the Environment (M. Betti/Action Leader)

L. Aldave de las Heras, M. Betti, M. Eriksson, G. Tamborini

Safeguards Research and Development (K. Mayer/Action Leader)

S. Abousahl, M. Betti, O. Cromboom, L. Duinslaeger, K. Lützenkirchen, K. Mayer, H. Ottmar, I. Ray, P. Richir, A. Schubert, G. Tamborini, P. van Belle

European Commission

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Abstract

The activity report 2004 of the Institute for Transuranium Elements (ITU) of the Joint Research Centre describes the progress made during the second year of the 6th Framework Programme for research of the European Union.

In the first part of the report an overview with a summary of the scientific objectives, important events, facts & figures and quality management of ITU is given.

In the second part of the report, a more in-depth technical description of progress made in the seven actions is given, including highlight articles on important progress made during 2004.

Topics covered focus on:

Basic Actinide Research

Single Crystal Growth of "115" Actinide Compounds

Spent Fuel Characterisation

Limits of the Radiolytic Enhancement of Spent Fuel Dissolution in Groundwater: Activity Dependence

Safety of Nuclear Fuels

Thermophysical Characterisation of UO₂ Fuel with a Burn-up up to 100 MWd/kgHM

Partitioning and Transmutation

First Americium Fuel Fabrication in the Minor Actinide Laboratory

Alpha-Immunotherapy

From Bench to Bedside: Targeted Alpha Therapy (TAT) for Malignant Melanoma

Measurement of Radioactivity in the Environment

Characterisation of Environmental U- and Pu-rich Particles by Synchrotron Radiation Based Techniques

Safeguards Research and Development

Characterisation of Radioactive Microparticles for Nuclear Safeguards Verifications and Detection of Undeclared Nuclear Activities

More information on the Institute and a list of previous Activity Reports may be found on the Internet:

<http://itu.jrc.cec.eu.int> and <http://www.jrc.cec.eu.int/>



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