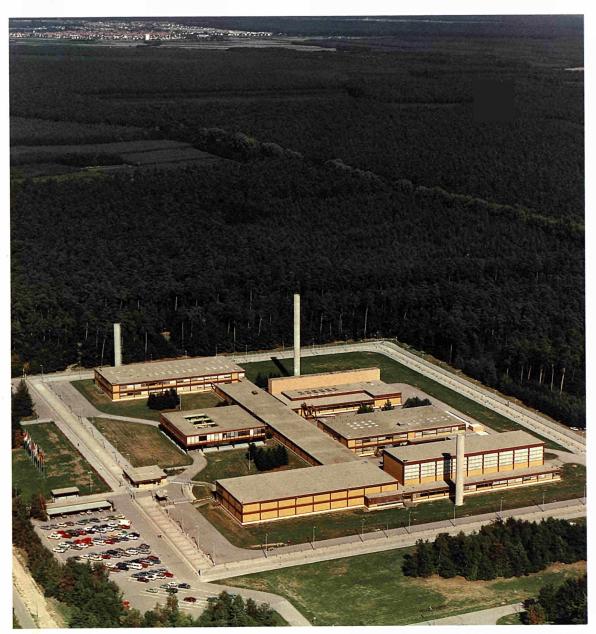
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Commission of the European Communities Joint Research Centre Karlsruhe Establishment



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European Institute for Transuranium Elements

Expertise and Programmes

COMMISSION OF THE EUROPEAN COMMUNITIES

THE EUROPEAN INSTITUTE FOR TRANSURANIUM ELEMENTS EXPERTISE AND PROGRAMMES

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J.-P.Contzen Director General JRC

Foreword

Looking forward

The 25th anniversary of the Institute for Transuranium Elements occurs at a time when new orientations have just been given to the Joint Research Centre after receiving the full support of each of the three institutions involved, Commission, Council and Parliament.

These new orientations do not fundamentally change the vocation or the mode of operation of the Institute, whose role has never been in question, and whose contribution to Community research has always been highly appreciated. Nevertheless, they will cast a new light on its activities.

One significant element is the concentration of JRC activities in general on issues related to Safety (nuclear safety as well as safety in conventional industrial sectors) and the Protection of the Environment. Within this framework, the Institute will continue to contribute to the improvement of safety at all stages of the nucleaf fuel cycle and should consider to what extent the knowledge so acquired could be used for other more general applications related to the protection of the environment.

Furthermore, the JRC is now entitled to devote a fraction of its financial resources to exploratory research, an activity which escapes multiannual programming and which is intended to maintain and enhance the scientific vitality of the Centre. This will enable the Institute, which has acquired a high reputation in the more fundamental aspects of actinide research, to explore new avenues of science.

At the other end of the spectrum of R & D activities, the JRC will increase its part of scientific and technical work performed under contract from third parties; this will be an opportunity for the Institute to market the specialised competences and facilities which it has developed over these past 25 years. Finally, there is a strong will to reinforce further the ties between the JRC Institutes and Scientific Institutions both in the Member States and at international level, notably through increased exchanges of grantholders and visiting scientists. This should strengthen the role of Karlsruhe as a reference point in the specialised field of transuranium elements.

The reform of the JRC now being implemented should give an added impetus to an Institute which has always shown a strong dynamism and an aspiration to scientific excellence.

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J.van Geel Director of the Institute

Introduction

In 1963, the cornerstone was laid for the European Institute for Transuranium Elements on the site of the German Nuclear Research Centre, following an agreement between the EURATOM Commission and the Government of the Federal Republic of Germany.

The mission of the Institute, as defined in the Annex of the EURATOM Treaty of 1957, concerns research and development work for improving the safety and the efficiency of the nuclear fuel cycle as well as fundamental studies on the physics and the chemistry of the transuranium elements.

Today, the Institute has some 200 employees, among them about fourty university-trained physicists, chemists, materials scientists, and engineers, assisted by highly specialised technicians and laboratory aids, as well as a number of office staff and administrators from eleven Member States of the Community. Presently, 15 visiting scientists and student trainees from various countries are working at the Institute.

Buildings and infrastructure are such that radioactive materials can be handled safely in special facilities. This puts the Institute in the rather unique position that a wide variety of activities ranging from fundamental and applied research to engineering studies can be executed "under one roof".

The organisation of the scientific-technical staff is such that it combines the advantages of high specialization with a wide degree of flexibility.

Practically all R&D work presently carried out at the Institute with a relation to nuclear energy is safety-oriented. Our principal themes for the 1988-91 programme are:

- The safety of nuclear fuels, in particular under transient and off-normal conditions and in advanced reactor concepts,

- The safety of the fuel cycle and its effect on the environment, comprising actinide formation and recycling studies and the investigation of nuclear aerosols,

- Basic actinide research, dealing with the preparation of well-defined samples and the investigation of their properties,

- Investigations relating to reactor safety analysis,

- The characterisation of nuclear waste forms,

- Safeguarding of fissile materials.

The results of the work performed at the Institute have been described in more than 1000 publications in scientific journals, as books, monographs, or reports and have been presented at roughly 1500 major conferences and meetings. Almost 100 patents have been filed by staff members.

The specific research programmes executed at the Institute for Transuranium Elements have been evaluated in the past by a Panel of independent specialists from various countries. In their report on 'The Evaluation of the JRC Programme on Nuclear Fuels and Actinide Research 1980/86', they come to the conclusion that the Institute is internationally recognized as a 'Centre of Excellence' for its studies of the physics and the chemistry of the heavy elements, and that the orientation of the Community programme on nuclear fuels and actinide research corresponds to current needs.

The following text gives an overview of programmes and prospects of the Institute, it describes expertise and equipment of the various scientific-technical units and summarises some of their recent achievements.

We invite interested parties, private industry or government institutions, to make use of the know-how and the facilities available at the Institute and collaborate with us for the benefit of the European citizen.

Requests for more information should be addressed to

The European Institute for Transuranium Elements Programme Office Postfach 2340 D-7500 Karlsruhe 1

Programmes and Prospects

The research and development work at the European Institute for Transuranium Elements is executed according to decisions of the Council of Ministers of the European Communities, which were taken, after consultation of the European Parliament, on the basis of proposals from the European Commission. These 'specific research programmes' are embedded in a long-term strategic plan of European research and development, the "Framework Programme". Besides specific research programmes, the Institute carries out research tasks upon request from other Commission services and performs contract work against payment for customers from industry or national institutions.

1. Specific Programmes

The research programmes carried out at the Institute for Transuranium Elements of the Joint Research Centre are dealing with the investigation of properties, handling risks and possibilities of technological application of the heaviest elements in the Periodic Table, elements, all of which are radioactive and some fissile (Fig. 1).

Research on radioactive material implies high cost, licensing problems and unusual safety precautions, which exclude universities and conventional national research institutions from entering into the field. It thus constitutes an excellent example for the beneficial effect of Community efforts providing funds and facilities which otherwise would not have been available.

The main part of the work performed at JRC Karlsruhe concerns research on nuclear fuels and fundamental studies on actinides. Other activities deal with problems of reactor safety, of fissile materials control and, to an important extent, with the management of nuclear waste. Relations exist with programmes on Environmental Research and on Materials Research. An important tool for the fuels part of the programme is the high-flux reactor at Petten, which offers a wide variety of means for testing and experimentation.

All these activities are in line with Annex I, relating to Article 4 of the Euratom Treaty, which lists, among other areas, the chemistry and metallurgy of plutonium and other transuranium elements, their applications and their biological effects as fields for which the Commission should provide means of Community research. The programmes are structured around four "themes", namely "Safety Studies for Nuclear Fuels", "Safety of the Fuel Cycle and its Effect on the Environment", "Actinides Research", and "Actinides Information Centre".

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Safety Studies for Nuclear Fuels encompass the optimisation of current and future power reactor fuels under safety aspects. Studies of the macro- and micro-scopic fuel structure under irradiation and corresponding model calculations render valuable information on the fission product behaviour which is the determining factor for fuel performance evaluation and risk assessment in case of core damage. These studies are complemented by measurements of thermodynamic and thermophysical fuel properties at normal fuel operating temperatures and above, for which unique expertise and equipment are available. While efforts to improve fuels for future fast breeder reactors will not be extended, the implications of using plutonium as fuel in today's light water reactors will merit closer attention. Studies on the radioactive inventory which might be released in case of an accident (source term) are being pursued in close collaboration with the Project PHEBUS PF and the JRC Project on Reactor Safety. A characterisation of debris of TMI-2 fuel is carried out at the Institute for Transuranium Elements in Karlsruhe in close contact with the OECD Nuclear Energy Agency.

Investigations related to the Safety of the Fuel Cycle and its Impact on the Environment will play an increasingly important rôle in the Institute's research programmes. They are a logical continuation of previous work aimed at reducing long-term radiological risks of nuclear energy production by studying in detail the formation of actinides and improving means of their analysis, and by dealing with selected topics to improve methods of reprocessing and recycling of nuclear fuels. Further efforts will be made to improve industrial safety and mitigate consequences of incidents by studying possibilities of release and controlled manipulation of nuclear (and otherwise toxic) aerosols. A spin-off of these activities, related to the scavenging of toxic aerosols, might have a bearing on the Programme on Industrial Hazards. The characterisation of nuclear waste forms which constitutes a large fraction of the activities of the Institute, is performed in the framework of the JRC Programme on the Management of Nuclear Waste. In this context, problems related to the final disposal of spent fuel rods without reprocessing will be studied in detail. Improved techniques for safeguarding nuclear materials are being developed at Karlsruhe under the auspices of the JRC Programme on the Management of Fissile Materials.

Actinide Research is primarily basic in nature, where the European Institute for Transuranium Elements plays a central rôle on the European scene and contributes significantly to a better understanding of the physical chemistry and the solid state physics of actinides and their compounds by producing highly pure, well characterised samples for solid state research, by using advanced experimental techniques like photo-electron spectroscopy and high-pressure Xray diffraction for physical investigations on actinides and by promoting progress in the theoretical understanding of the electronic structure of the heavy elements. Directing this fundamental work towards problems like magnetism and superconductivity might help to understand phenomena of eminent technical significance. Actinide research is executed in a wide frame of international collaboration comprising teams from more than 40 European laboratories.

Actinides Information Centre: Efforts are continued to systematically collect and keep readily available (possibly as a public service) bibliographical information and physical, chemical and radiological data related to actinides and transuranium elements so as to make the Transuranium Institute at Karlsruhe a European information center on the properties of heavy elements and their compounds. Close relations are being maintained with actual and potential users of the results obtained in the frame of these programmes. This are on one hand the nuclear industry and the utilities, and, on the other hand, regulatory bodies, dealing with industrial safety.

2. Work for Commission Services

On behalf of the European Commission's Directorate-General of Energy, Directorate of EURATOM Safeguards, the Institute analyses samples which have been collected by EURATOM inspectors from nuclear plants in Europe under the international non-proliferation treaty in order to determine the nature and the amount of fissile materials transfered to and from these installations. Highly sophisticated equipment is used for the collection, preparation and analysis of these samples and most of the analytical operations are performed in a computercontrolled automatic laboratory.

3. Work for Third Parties

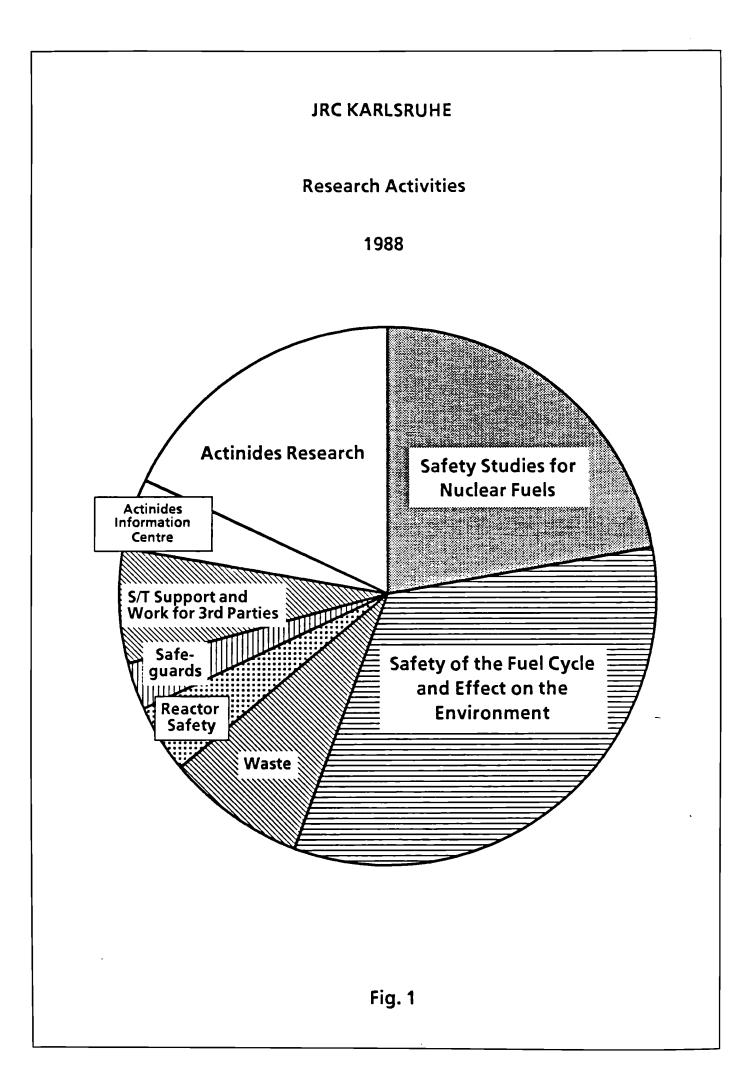
Contract work is being performed at the Institute in the fields of highperformance nuclear fuel analysis, preparation of actinide-containing compounds for testing and standardisation, fuel rod modelling and conditioning of waste, which is difficult to treat in large-scale commercial installations. The Institute performs post-irradiation analysis of plutonium-containing light-water reactor fuels, measures the fracture toughness of ceramics, and determines the oxygen potential of oxide fuel materials before and after irradiation upon request from third parties. A major multiannual contract concerns the preparation and characterisation of alloys containing "Minor Actinides" which might be of interest for a novel reactor concept, aiming at the conversion of long-lived nuclear waste constituents into short-lived fission products.

4. Exploratory Research

A small percentage of the Institute's potential is devoted to the exploration of new fields of research and development which would profit from the available expertise, but are not covered by the ongoing programme. The acceleration of aerosol scavenging under the effect of an acoustic field is such an area, to which considerable efforts are presently being devoted.

The programmes described above are flexible in the sense that the proportion of safety-related work to activities with an accent on nuclear technology can easily be adjusted to arising needs. Efforts to relate the results of fundamental studies directly to problems of technological relevance will be stepped up.

All R&D activities presently executed at the Institute are listed in Table 1.



European Institute for Transuranium Elements Programme Structure 1988/89 (Status October 1988)

A. Specific Programmes

100 Nuclear Fuels and Actinide Research

110 Safety Studies for Nuclear Fuels
111 Oxide Fuel Transients
112 High Burn-Up Chemistry
113 Optimisation of Dense Fuels
114 High-Temperature Properties of Nuclear Fuels

120 Safety of the Fuel Cycle and its Effect on the Environment
121 Formation of Actinides In-Pile
122 Safe Handling of Nuclear Fuels
123 Reprocessing of Nuclear Tuels

130 Actinides Research
131 Preparation, Characterisation and Application of Actinides
132 Study of the Electronic Structure of Actinides
133 High-Pressure Studies on Actinides

140 Actinides Information Centre

200 Reactor Safety

210 Fuel Behaviour under Accident Conditions

300 Radioactive Waste Management

310 Nuclear Waste Characterisation

400 Safeguards and Fissile Materials Management

410 R&D in Destructive Analysis of Fissile Materials

B. Work for Third Parties

B1.Scientific-Technical Support to Commission Services

701 Evaluation, Automation of Analytical Techniques for DG I 702 Analytical Support to DG XVII

B2. Contract Work for External Customers

C. Exploratory Research

Tab.1

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Expertise and Equipment

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J. Fuger

Chemistry Division

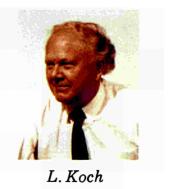
The activities of the Chemistry Division are dealing with the basic and applied research on the nuclear fuel cycle in a broad sense.

In particular, the properties of the actinide elements in relation with the aqueous reprocessing have been extensively studied with a special emphasis on separation methods, and the head-end aspects of the aqueous reprocessing of advanced fuels (carbides, nitrides) have been examined. Problems related to the presence of the actinides in waste solutions and waste forms (glasses, ceramics, spent fuels) have also been tackled, and studies aimed at the elimination of the actinides by partitioning and nuclear transmutation are of current interest.

The understanding of the properties of the actinides and their compounds in terms of their electronic structure is also a prominent area of study. As a prerequisite to such studies, methods for the preparation of radioactive samples of high purity, mostly in the form of single crystals, have been developed, together with characterization techniques by X-ray diffraction and chemical analysis. Although most samples are produced and characterized in our Institute, this project involves extensive collaborations with many other Laboratories which have equipment and expertise complementary to ours.

Experimental techniques used to study the electronic structure of the actinides and their compounds involve X-ray diffraction, optical reflectivity and resistivity under high pressure, ultra violet and X-ray photoelectron spectroscopy, elastic and inelastic neutron scattering, magnetic susceptibility to cite only a few. All these studies are carried out in close interaction with theoreticians specialized in band structure calculations.

The determinations of the chemical composition and isotope ratios in nuclear fuels samples before and after reactor irradiation and in reprocessing solutions is also one of our very prominent tasks. In that context are performed safeguards analyses on behalf of the EURATOM Directorate for Fissile Materials Control (Luxembourg). Another important activity is the development and the adaptation to glove box and hot cell work of computer controlled automatic analytical equipment and of expert systems to supervise and control the analytical operations.



Analytical and Radiochemistry

Competences

The analytical capacity is focused on (a) the determination of the chemical composition of and trace impurities in fuels and (b) on the analysis of isotope concentrations or ratios in fuel samples before and after irradiation in a test or power reactor as well as in samples taken from reprocessing plants.

The analytical laboratory has to cope with a complex variety of requests from the basic science sections as well as from the fuel developing and testing laboratories and disposes of a wide range of techniques, some of which had to be especially developed or adapted to be used with radioactive substances. Since it has routine work to do, most of the equipment is fully automated and provided with computerised data acquisition and evaluation systems.

One of the tasks of the radiochemical laboratory is to study the build-up of actinides during irradiation of uranium- and plutonium-fuels in a reactor. Besides this, the Group develops and tests methods to be used for fissile materials control in nuclear installations and, on behalf of the Commission's Directorate of EURATOM Safeguards, performs isotope analyses on samples (more than 1000 per year) which have been taken by EURATOM inspectors from nuclear plants in the frame of the international non-proliferation treaty. Most of the preparatory work is done in a computer-controlled automatic laboratory (Fig.1). Robot systems have been adapted for work in glove boxes, so reducing the risk of operators being exposed to radiation. Actually, the use of an expert system to supervise and control the analytical operations is under study.

The laboratory prepares and supervises activities aimed at transmuting longlived actinides from nuclear waste into short-lived fission products. In particular, it has launched in this context an irradiation of "Minor Actinide" (i.e. Np, Am, Cm)-containing oxide fuels in the French PHENIX reactor and is responsible for the execution of a contract concerning the preparation and characterisation of "Minor Actinide" alloys for a novel reactor concept.



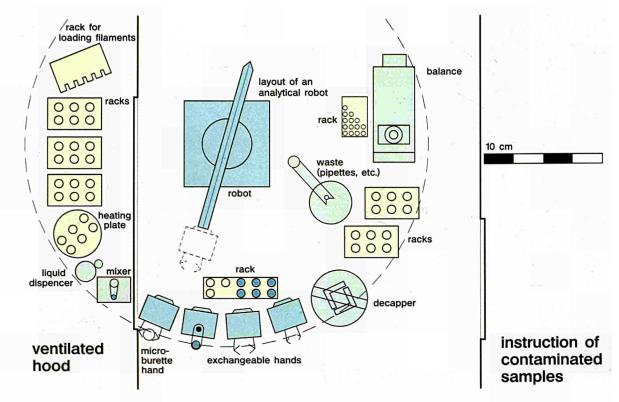


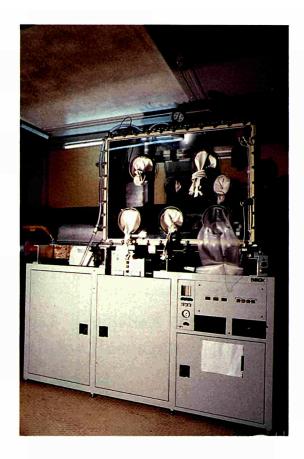
Fig. 1 Layout of on-site robotized reprocessing input analysis

Important Equipment

- Thermal ionisation mass spectrometers;
- Gas mass spectrometers;
- Inductively coupled plasma source mass spectrometer (ICPMS) (Fig. 2);
- Atomic absorption spectrometer;
- Laboratory robots;
- 8-way alpha counters;
- Gamma spectrometers with germanium detectors;
- K-edge densitometer;
- Inductively coupled plasma source atomic emission spectrometer;
- X-ray fluorescence equipment;
- Automatic titrator.

Principal Achievements During the Last Three Years

- Complete destructive analysis of the nuclide inventory of LWR fuels;
- Establishment of a set of integral cross sections for a fast reactor;
- Analysis of leachates from leaching studies of nuclear waste forms;
- Design of a Minor Actinide containing fast reactor fuel;
- Irradiation of Minor Actinide fuel in KNKII and PHENIX (Superfact);
- Measurement of isotopic abundances in Minor Actinide containing samples after irradiation in the KNK II reactor;
- Development of new methods for Np analysis;
- Development of a new, rapid mass spectrometric measurement technique for sub-nanogram amounts of U and Pu;
- Adaptation of laboratory robots for work with radioactive material;
- Adaption of ICPMS equipment to hot cell environment;
- Development of a concept for on-site nuclear material safeguards analysis at the input of a reprocessing plant.



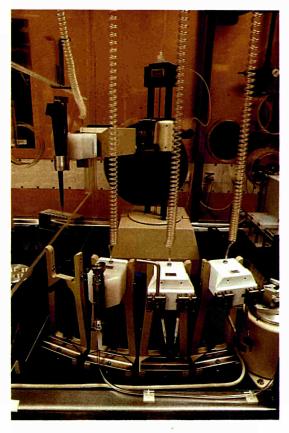


Fig.2 Inductively coupled plasma mass spectrometer

Fig.3 Laboratory robot in glove box

Some Recent Publications*

T.Adachi, K.Kammerichs, L.Koch, Inherent Isotope Dilution Analysis of ²³⁷Np in Spent Nuclear Fuels, J.Radioanal.Nucl.Chem.Letters 117(4)(1987) 233

L.Koch, New Procedures to Verify the Input Analysis of Nuclear Fuel Reprocessing in vol.4, Nukleare Entsorgung, VCH-Verlag, Weinheim 1988 M.Romkowski, S.Franzini, L.Koch, Mass Spectrometric Analysis of Sub-

M.Romkowski, S.Franzini, L.Koch, Mass Spectrometric Analysis of Sub-Nanocurie Samples of Uranium and Plutonium, 9th Annual Meeting of the European Safeguards Research and Development Association, 12 - 14 May 1987, London, GB

K.Mayer, L.Koch, J.Naegele, K.Richter, On the Interaction of Sodium with Hypostoichiometric Uranium-50% Americium Mixed Oxides, 18èmes Journées des Actinides, 20 - 22 April 1988, Paris, France

O.Cromboom, H.Kutter, M.Ougier, C.Apostolidis, H.Bokelund, Determination of Traces of Thorium and Neptunium in Plutonium Nitrate Solutions, 17èmes Journées des Actinides, 26 - 28 March 1987, Lausanne. Switzerland

* further relevant references may be found in "JRC Karlsruhe - List of Publications 1986/87" (EUR-Report 11790 EN, 1988)



G.H. Lander

Actinide Research

Competences

The Actinide Research Section is concerned essentially with fundamental studies, aiming at producing new actinide compounds, usually in the form of single crystals, and measuring and understanding their properties.

The effectiveness of the Section is greatly increased by an extensive collaboration with many other laboratories, which have equipment and expertise complementary to that found at Karlsruhe. Most of these external laboratories use for their studies samples which have been produced and characterised by the Karlsruhe Group.

In its Research Evaluation Report^{*}, a panel of independent experts states, that "The actinide research programme at JRC Karlsruhe ...makes an important contribution to research in materials science throughout the European Community. During recent years, a number of very significant discoveries concerning the physics and chemistry of the actinides have been made, and Karlsruhe is internationally recognized as a 'Centre of Excellence' for actinide research".

Thus, expertise is available for the preparation of radioactive, mostly single crystalline, samples with high purity, and for their characterisation by analytical and x-ray diffraction techniques.

Experimental techniques used to obtain information on the electronic structure of actinides and actinide compounds comprise elastic and inelastic neutron scattering as well as ultraviolet and x-ray photoelectron spectroscopy.

A team of theoreticians is specialised in band structure calculations.

*Evaluation of the JRC Programme on Nuclear Fuels and Actinide Research 1980-1986, Research Evaluation Report No.26, Commission of the European Communities, DG XII/JRC, Brussels 1986

Important Equipment

- Furnaces for preparing actinide alloys and compounds;
- Installations for the preparation of single crystals by various techniques;
- A photoemission electron spectrometer with ultraviolet and x-ray excitation for use with alpha active samples;
- X-ray diffraction equipment for the determination of crystal structures and lattice parameters



Fig.1

Picture of single crystal (5mm on edge) of PuSb, made from the rare isotope ²⁴²Pu. These crystals can be cooled to low temperatures because there is no self-heating from the ²³⁹Pu nucleus and are therefore ideal for resistivity and magnetisation experiments. In addition, ²⁴²Pu does not absorb thermal neutrons like ²³⁹Pu, so these crystals can also be used in experiments involving the scattering of thermal neutrons. (The ²⁴²Pu isotope was supplied by the US Department of Energy).

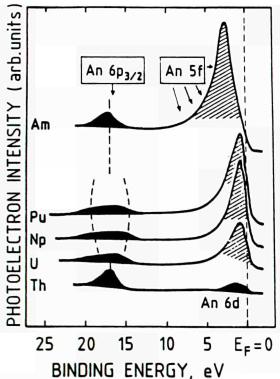


Fig.2

X-ray photoemission spectra of the actinide metals. Of particular interest is the intensity and the position of the 5f emission. In Th there are no 5f states. In U, Np, and Pu the 5f states lie very close to the Fermi level E_F , this means these electrons strongly interact with their neighbours and the elements are highly reactive. In Am, on the other hand, the 5f level has dropped below E_F , and Am can be regarded in this sens as a localized (lanthanide-like) element.

Principal Achievements During the Last Three Years

- Preparation of single crystals of Pu and Np pnictides and chalcogenides;
- Preparation of single crystals of ²⁴²PuSb for specific heat, magnetisation, and neutron scattering experiments. Discovery of an enormous magnetic anisotropy in this Pu system (Fig. 1),
- Preparation of single crystals of NpRu₂Si₂ for resistivity, specific heat, and neutron scattering experiments;

- Mössbauer experiments on NpSb-NpTe solid solutions to show charge transfer effects;
- Photoemission experiments on Am and Pu to show localised-delocalised transition (Fig. 2);
- Photoemission experiments on U-Ni surfaces to investigate catalytic properties;
- Neutron diffraction experiments to show covalency in UCl₄;
- Polarized neutron experiments to show anisotropy effects in PuFe₂;
 Theoretical (single-electron band structure) prediction of a new type of magnetism in which the orbital and spin moments cancel. These predictions have now been verified in actinide Laves phases (PuFe₂ and UFe₂) by polarized neutron diffraction.

Some Recent Publications*

J.W.Ward, W.Bartscher, J.Rebizant, A Re-Examination of the Neptunium-Hydrogen System, J.Less-Common Met. 130 (1987) 431

J.C.Spirlet, J.R.Peterson, L.B.Asprey, Preparation and Purification of Actinide Metals, Adv.Inorg.Chem. and Radiochemistry 31(1987)1

P.Burlet, J.Rossat-Mignod, G.H.Lander, J.C.Spirlet, J.Rebizant, O.Vogt, Neutron Diffraction Study of PuSb: The Critical Regime, Phys.Rev.**B36** (1987) 5306

A.J.Freeman, G.H.Lander (Eds.), Handbook on the Physics and Chemistry of the Actinides, North Holland Publ.Co. (1987)

P.A.Thiry, J.J.Pireaux, R.Gaudano, J.R.Naegele, J.Rebizant, J.C.Spirlet, Infrared Dielectric Response of UO₂ Single Crystal Surfaces Investigated by High Resolution Electron Energy Spectroscopy, J.Chem.Soc.(Faraday Transactions II) 83 (1987) 1229

M.S.S.Brooks, O.Eriksson, B.Johansson, Spin-Orbit Effects in the Energy Bands of Actinide Transition Metal Intermetallics, 17èmes Journées des Actinides, 26 - 28 March 1987, Lausanne, Switzerland

J.Fuger, Actinide Research at the European Institute for Transuranium Elements, 2nd Int.Conf. on the Basic and Applied Chemistry of f-Transition and Related Elements, 6 - 10 June 1987, Lisbon, Portugal

* further relevant references may be found in "JRC Karlsruhe - List of Publications 1986/87" (EUR-Report 11790 EN, 1988)



High-Pressure Studies

Competences

For almost ten years, the property changes of actinides and actinide compounds under the effect of pressure are studied at the European Institute for Transuranium Elements. These investigations have turned out to be a useful and even necessary part of actinide solid state research, since they contribute to the understanding of the electronic structure of these materials. The results of this work have attracted international attention, and the High-Pressure Studies Section of the Institute is considered to be one of the leading institutions in the field.

High pressure is generated by means of diamond anvil cells (DAC) (Fig. 1), which in some of the most advanced versions can generate pressures over 2 Mbar. Applications range from solid state studies to geophysical research, and some of the properties of which the pressure dependence is determined in a DAC are crystal structure, compressibility, electrical conductivity, and optical properties.

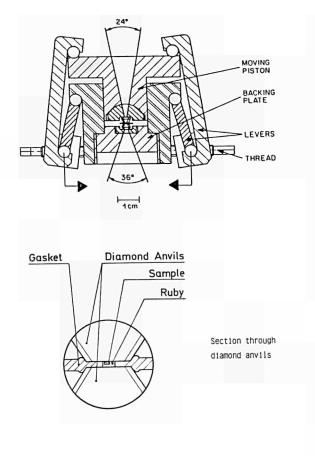
Important Equipment

- Diamond anvil cells for x-ray diffraction, x-ray absorption, and optical studies of materials under pressures up to 600 kbar;
- Equipment for energy-dispersive x-ray diffraction and for angle-dispersive x-ray diffraction with a position-sensitive detector;
- Equipment for the measurement of optical reflectivity and optical absorption under pressure;
- Equipment for the measurement of electrical resistivity under pressure (under development);
- Equipment for x-ray absorption and x-ray diffraction studies under pressure to be used at the synchrotron storage rings of DESY (Hamburg) and LURE (Orsay).

Principal Achievements During the Last Three Years

X-ray diffraction and absorption measurements at high pressure have been performed with a large number of actinide metals and compounds. The nature of the high pressure phases observed, the volume changes measured at the phase transition, the compressibilities determined as well as the observed variation of the absorption edge with pressure are important elements for the determination of the rôle of the 5f electrons in these materials.

The complete high-pressure structural systematics of the accessible heavy actinide metals (Am, Cm, Bk, Cf) was established. All four metals undergo phase transformations under pressure, where a transition to a low-symmetry (orthorhombic) structure corresponds to 5f delocalisation (Fig. 2). The most remarkable member of this group is the 5f7-configurated curium: It needs the highest pressure to undergo transformation and exhibits the largest density increase upon delocalisation, which makes it particularly interesting for further studies.



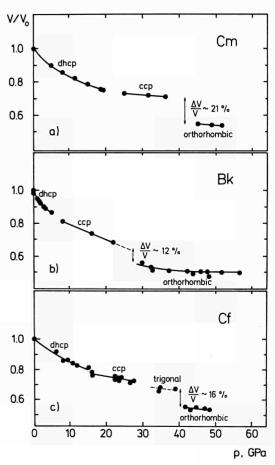


Fig. 1

The Syassen-Holzapfel high-pressure cell. The knee-type lever arms generate the thrust for the piston when the threads are advanced synchronously with a gear-set wrench. Fig. 2

Pressure-volume relationships of Cm, Bk, and Cf. $\Delta V/V$: volume decrease upon delocalisation of the 5f electrons.

For the first time, high-pressure investigations were extended to interactinide alloys: Two perfectly homogeneous Bk-Cf alloys were studied by high-pressure xray diffraction. The transition pressures, the structural behaviour, and the density change upon delocalisation were found to vary smoothly with composition.

From these and the previous studies of actinide metals, a general diagram of the phase relations of actinide metals under pressure was established (Fig. 3) This diagram allows us to recognize trends in structures and phase transitions and to determine where further work in the field is most desirable.

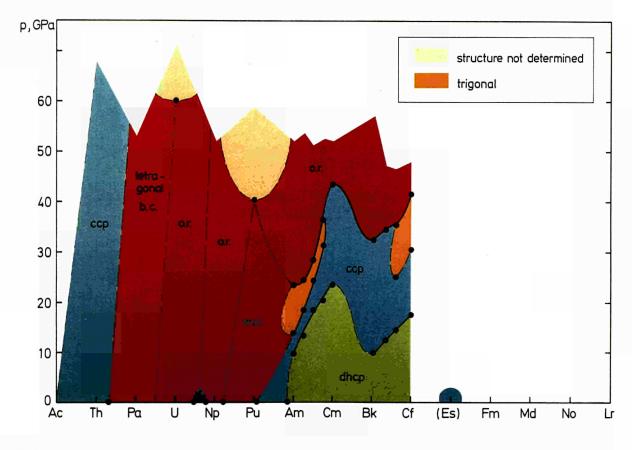


Fig.3

General diagram of the phase relations of the actinide metals under pressure. Increasing pressure, as well as decreasing atomic number, both transform high-symmetry crystal structures into structures of lower symmetry: double hexagonal close packed (green area) \rightarrow cubic close packed (blue area) \rightarrow orthorhombic and monoclinic structures (red area).

The high-pressure behaviour of a large number of NaCl-type actinide compounds was investigated. Most of them transform to the CsCl-structure type. The fact that ThC and ThN (which have no occupied 5f levels) do not exhibit a phase transition under pressure indicates that the 5f electrons are involved at least in some of the phase transitions of the other actinide compounds.

With the help of absorption spectroscopy, phase transformations with pressure were identified for the first time in $CfBr_3$ and in AmI_3 . The measurement of the optical reflectivity of UO_2 under pressure made it possible to follow the narrowing of the f-d gap with increasing pressure and showed that pressure induces f-d mixing even before the structural transition occurs. In NaCl-type actinide compounds which exhibit a transition to the CsCl structure type under pressure, this transition is accompanied by the disappearance of reflectivity bands.

Part of these results were obtained in collaboration with external laboratories, for example the Oak Ridge National Laboratory, the University of Copenhagen, the Institut Curie, and the Max-Planck-Institut für Festkörperforschung.

Some Recent Publications*

U.Benedict, Structural Data of the Actinide Elements and of their Binary Compounds with Non-Metallic Elements, J.Less-Common Met. 128(1987)7

U.Benedict, The Effect of High Pressure on Actinide Metals, in volume 5 of the Handbook on the Physics and Chemistry of the Actinides (A.J.Freeman and G.H.Lander, Eds.), North Holland Publ.Co. 1987

U.Benedict, General Diagram of the Phase Relations of Actinide Metals under Pressure, High Pressure Research (subm.)

H.Luo, Ś.Dabos, U.Benedict, J.C.Spirlet, Compression of the Heavy-Fermion Compound URu₂Si₂ to 50 GPa, J.Less-Common Met. (subm.)

* further relevant references may be found in "JRC Karlsruhe - List of Publications 1986/87" (EUR-Report 11790 EN, 1988)



H. Bokelund

Chemical Technology

Competences

The Chemical Technology Section is concerned with the chemistry of the actinides under technological aspects, i.e. the reprocessing of nuclear fuels; the separation and chemical purification of actinides by precipitation, ion exchange, solvent extraction, high-pressure liquid chromatography, and extraction chromatography; the characterisation of waste forms (high and intermediate level waste); the treatment of alpha waste; photochemical methods; radiochemical analysis, spectroscopy, speciation analysis, gas analysis, neutron activation analysis; and automated data acquisition and handling.

Important Equipment

- Spectrophotometer with an on-line computing system;
- Optical multichannel analyser;
- Alpha- and gamma-spectrometers, comprising detectors for the low energy range (Fig. 1);
- Gas analysers;
- Equipment for high-pressure liquid chromatography;
- Ion chromatograph.

Principal Achievements During the Last Three Years

- Elaboration of head-end schemes for the reprocessing of advanced fuels;
- Determination of the compositions of off-gases from the dissolution of oxides, nitrides, and carbides (Fig. 2);
- Investigations into the use of ¹⁵N in a (U,Pu)-nitride fuel cycle.



Fig.1 Measurement station for y - spectrometry

- Study of redox properties of Pu and Np and of Pu colloïds;
- Design of a separation scheme for actinides from fuel and waste solutions (Multipurpose Unit);
- Systematic study of the absorption spectra of actinides;
- Removal of actinides from intermediate waste and characterisation of the final waste form (cement matrix);
- Statistical evaluation of leaching procedure on glass.
- Implementation of leaching tests on active glasses in a hot cell;
- Participation in interlaboratory simulation tests on repository systems organized by the Commission of the European Communities.

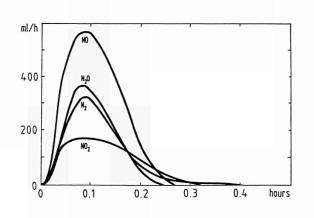


Fig.2 Gas evolution during dissolution of UN-specimens

Some Recent Publications*

H.Bokelund, K.Deelstra, Statistical Tests Applied as Quality Control Measures to Leaching of Nuclear Waste Glasses and in the Evaluation of the Leach Vessel, Radioactive Waste Management and the Nuclear Fuel Cycle, 11 (1988) 61

C.Apostolidis, H.Bokelund, A,Moens, M.Ougier, Redox Behaviour of Neptunium in the Presence of U, Pu, and Am, Applied in their Separation by TBP and Ion Exchange, Inorg.Chim.Acta 140(1987) 253

J.P.Glatz, H.Bokelund, S.Zierfuss, Dissolution of Different Types of Nuclear Fuels in Nitric Acid, International Conference on Nuclear Fuel Reprocessing and Waste Management, August 1987, Paris, France.

C.Apostolidis, H.Bokelund, M.Ougier, Speciation of Neptunium in Concentrated Nitric Acid; Absorption Spectra of Np(IV), Np(V), and Np(VI), 18e Journées des Actinides, 20 - 22 April 1988, Paris, France

* further relevant references may be found in "JRC Karlsruhe - List of Publications 1986/87" (EUR-Report 11790 EN, 1988)



H.Blank

Physics Division

The Physics Division is concerned with problems of materials science and thermodynamics and comprises laboratories which are carrying out studies on nuclear fuel development, characterisation, and testing, with an accent on safety aspects.

In particular, phase diagrammes of uranium and plutonium bearing compounds, vapour pressures (up to 6000 K) and optical emissivity around the melting point, physical properties like thermal conductivity and optical absorption are being studied. Another group of topics deals with structural and irradiation induced lattice defects, mechanical properties, and diffusion. These studies serve to initiate the fabrication and irradiation of new fuels in thermal and fast reactors. After irradiation, these fuels are examined in our hot cell laboratory. Thus, a close collaboration of the laboratories of the Physics Division performing basic research with the fuel fabrication group and the hot cell laboratory is always mandatory.

The results thus obtained provided the basic data and the information necessary for the interpretation and the modelling of the in-pile fuel performance.

Up to the beginning of the eighties, the laboratories of the Physics Division were carrying out mainly basic and applied research for the development and testing of new fast reactor fuels, such as mixed U, Pu-oxides, and to a large extent mixed carbides, carbonitrides and nitrides.

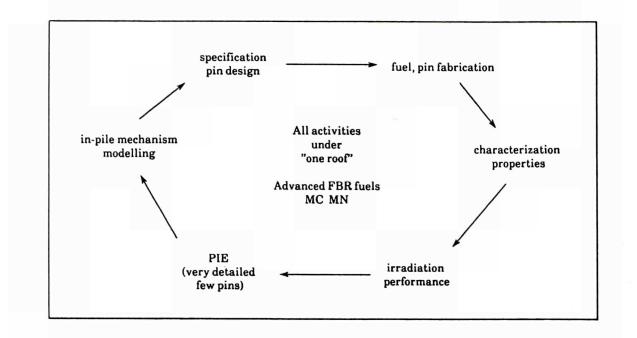
All activities were guided by two principles:

- i) to close the gap between basic material science and nuclear fuel technology and applications;
- ii) to promote the feed-back cycle which is necessary for the efficient development of any new fuel with increased safety features and highly reliable inpile performance (see the figure below).

The Institute is in the very fortunate position that all activities involved in this feed-back cycle (with the exception of the reactor irradiations) can be carried out

under one roof. Thus the required flux of information between the various stations in the cycle is easy to maintain.

This fuel research is performed in close contact with the corresponding national research laboratories in the European Community.



Feed-back cycle for the development of new nuclear fuel materials with increased safety properties and highly reliable in-pile performance

In the last 8 years, new problems came up and had to be solved by the laboratories of the Physics Division, in response to changing demands from the public and the industrial nuclear sector. The fast reactor fuel activities were strongly reduced and concentrated on nitride fuel development. The analysis of LWR fuel irradiated to high burn-up was carried out under the aspect of safety and reliability in collaboration with industry and international research projects. Furthermore, the know-how and methods developed during the studies of ceramic nuclear fuels are now being applied to the investigation of the properties of highlevel waste forms, such as glass, synrock, and spent fuel.

A lively new project within the general frame of the safety of the nuclear fuel cycle deals with the study of the properties of Pu-bearing aerosols. Recently, this project was extended to an investigation of the possibilities of scavenging of aerosols by subjecting them to a high-intensity acoustic field. This idea came up in our high-temperature laboratory after the successful use of acoustic levitation of specimens for the contactless measurement or optical emissivity, thermal expansion and heat capacity of UO_2 around the melting point.

The combination of basic materials science and physics with technological applications, as it has been practised over the past 25 years in this Division, is a permanent challenge to its Scientists who must learn anew for each problem to look "both ways". However, this working philosophy will guarantee success, also in the future.



C.Ronchi

High Temperature Property Studies

Competences

The Group has been formed by combining the laboratories for Thermodynamics and for Thermophysical Property Studies, both with an outstanding international reputation in their respective fields. The team is composed of chemists, physicists and specialists in materials science. Special technical competences comprise electronics, laser technology, mechanical engineering, computerized data acquisition techniques, and numerical analysis.

The Group is presently involved in investigations ranging from basic thermodynamics (equation of state of nuclear fuels, measurement of equilibrium vapour pressures, enthalpy, and specific heat), energy transport phenomena (measurement of the thermal conductivity of solid and liquid fuels, thermal conductivity of irradiated fuel materials) to more engineering-oriented problems like the study of the vaporization of fission products from irradiated nuclear fuels.

A field of special interest in this context is the study of radiative properties of refractory materials at high temperatures, for which advanced pyrometric methods have been developed.

Pioneering work has been done in establishing "clean" conditions for experimentation at extremely high temperatures: A recent development is the combination of laser-pulse heating with acoustic levitation.

Important Equipment

- A high-temperature research facility consisting of a modular cell in which temperatures up to 7000 K can be reached by laser pulse heating. The system is equipped for analysing the evaporated species by mass spectrometry, highspeed photographing of emitted shock-waves, performing ion density measurements and measuring spectral temperatures and evaporation rates (Fig. 1).

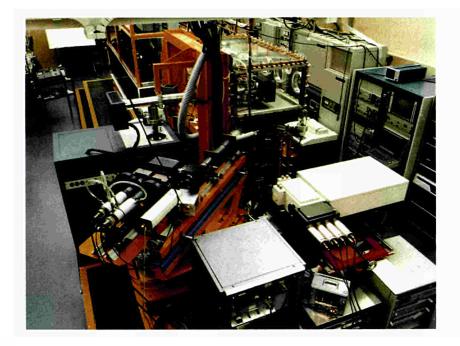


Fig. 1

General view of equipment for high temperature vapour pressure studies with laser heating, high-speed pyrometry and mass spectrometric analysis of the evaporated species

- An autoclave equipped with an ultrasonic levitator for laser-pulse heating of freely suspended samples under buffer gas pressures of up to 4 kbar to extremely high temperatures. The sample temperature is measured with a high-speed six-colour pyrometer developed for this purpose. Variations of sample dimensions during heating and cooling can be observed by an x-ray shadow casting technique (under development). The apparatus is presently being used for the study of calorimetric and radiative properties of materials up to 5000 K. (Fig.2)
- A pressurized furnace in which samples can be heated by two continuous highpower lasers, equipped with a high-precison temperature scanning system to record the 2D-temperature profiles on the faces of the tablet-shaped sample. The device has been successfully used to measure the thermal conductivity of the liquid phase of ceramic nuclear fuel.
- Two installations to measure the thermal diffusivity of solid samples up to 2000 K based on periodic electron beam and laser-flash heating, respectively.
- Knudsen cell furnaces with mass spectrometers for the determination of equilibrium vapour pressures up to 3100 K.
- High-speed six-colour pyrometers for the simultaneous measurement of temperature and spectral emissivity on a millisecond time scale.

Principal Achievements During the Last Three Years

- Experimental determinations of the equilibrium vapour pressure over UO_2 were carried out up to 5000 K. Theoretical studies on the vaporisation mechanisms and on the high-temperature thermodynamics of UO_2 have

significantly contributed to the understanding of the equation of state of oxide fuels and thus to our present knowledge of the fuel behaviour under the conditions of hypothetical core-disruptive accidents.

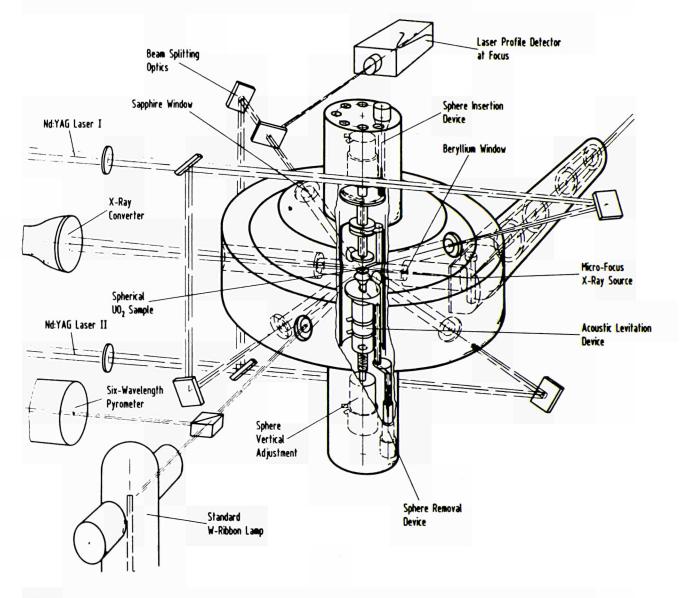


Fig. 2

Perspective drawing of tetrahedal laser heating of a spherical sample within a high-pressure autoclave, showing means for acoustic levitation, pyrometry, and x-ray shadow casting technique

- With recent measurements of the thermal conductivity of liquid UO_2 in this laboratory, a long-standing controversy related to the post-accident heat removal from a molten reactor core could be settled (Fig. 3).
- Novel calorimetric techniques have been developed, enabling one to measure the specific heat capacity of small samples at 5000 K and above.
- The spectral emissivity of refractory metals was measured up to and above their melting point. It was observed that the materials behave upon melting like grey bodies in the pyrometrically relevant wavelength range between 400 and 1000 nm.

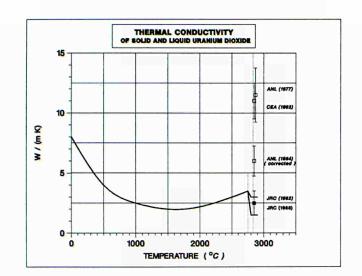


Fig. 3

The thermal conductivity of solid and liquid UO_2 . Comparison of ITU results with literature values above the melting temperature

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J.Magill, G.J.Hyland, M.H.Rand, New Ionic Contributions to the Vapour Pressure of Urania, J.Chem.Phys. (subm.)

J.Magill, R.Beukers, F.Capone, W.Heinz, J.-P.Hiernaut, E.Kramer, M.Martellenghi, R.Selfslag, P.Werner, A New Technique for Containerless Processing of Materials at High Temperatures, 5th Int.Conf. on High Temperature and Energy Related Materials, 25 - 29 May 1987, Rome, Italy

H.A.Tasman, Thermal Conductivity of Liquid UO₂, 11th Europ.Conf. on Thermophysical Properties, 13 - 16 June 1988, Umea, Sweden

H.E.Schmidt, R.W.Ohse, Thermophysical Properties Research on Nuclear Fuels at the European Institute for Transuranium Elements, 1st Asian Conf. on Thermophysical Properties, 21 - 24 April 1986, Beijing, China

J.P.Hiernaut, C.Ronchi, Use of Acoustic Levitation in High-Temperature Heating Experiments, 11th Europ.Conf. on Thermophysical Properties, Umea, Sweden, 13-16 June 1988

J.P.Hiernaut, F.Sakuma, C.Ronchi, Determination of the Melting Point and of the Emissivity of Refractory Metals with a Six-Wavelength Pyrometer, 11th Europ.Conf. on Thermophysical Properties, Umea, Sweden, 13-16 June 1988

R.W.Ohse, Extension of Thermophysical Property Measurements of Ceramic Materials up to 5000 K,10th Symposium on Thermophysical properties, Gaithersburg, Md, 20-23 June 1988

* further relevant references may be found in "JRC Karlsruhe - List of Publications 1986/87" (EUR-Report 11790 EN, 1988)



Fuel Characterisation

Competences

Metallic and ceramic nuclear fuels are characterised by their chemical composition and their structural features. Both may change when the material undergoes homogeneous changes in temperature or when applying thermal gradients across the specimen. The latter situation is especially interesting in view of the thermal conditions prevailing in-pile in an operating fuel rod, where radial temperature gradients of up to 20 000 K/cm may give rise to drastic changes in structure and composition.

The Fuel Characterisation Laboratory is specialized in the metallographic and ceramographic investigation of un-irradiated alpha-active materials by employing up-to-date techniques of microscopy and image analysis.

The laboratory is further equipped for the study of equilibrium thermodynamic properties of nuclear fuels and actinide compounds in general, with emphasis on phase diagram work, as well as for the investigation of reaction kinetics.

A particular competence is the study of the behaviour of nuclear fuel materials under heating conditions similar to those existing in a nuclear reactor (Fig.1).

Recently, an investigation of the thermodynamics and of phase relationships in Minor-Actinide-containing U,Pu,Zr-alloys was started.

Important Equipment

- Optical and scanning electron microscopes;
- Electronic image analyser (Quantimet);
- Microdensitometer;
- Microporosimeter;
- Dilatometer;
- Thermobalance;

- Equipment for differential thermal analysis;
 High-temperature press;
- Direct electrical heating apparatus.

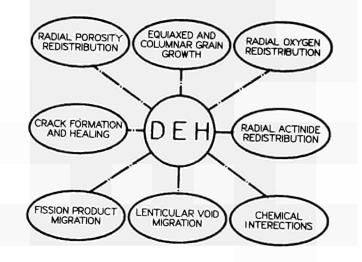
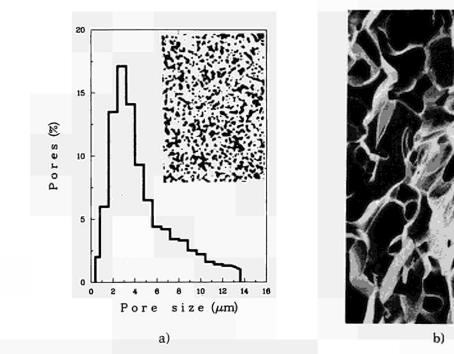
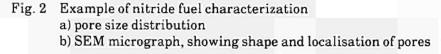


Fig. 1

Processes investigated by heating oxide , nitride and carbide fuel pellets in radial temperature gradients using the Direct Electrical Heating (DEH) apparatus





Principal Achievements During the Last Three Years

- Characterisation of nitride fuels for irradiation experiments NILOC and NIMPHE (Fig. 2);
- Investigation of the kinetics of the carbothermic nitriding reaction $UO_2 + PuO_2 + C + N_2$;
- Determination of the oxygen potential of hypostoichiometric neptunium oxide;
- Study of the grain growth kinetics of (U,Pu) mixed oxide fuel;
- Investigation of the baviour of U,Pu-nitride fuel (NIMPHE) in a temperature gradient;
- Study of the Neptunium-Oxygen phase diagram (Fig. 3).

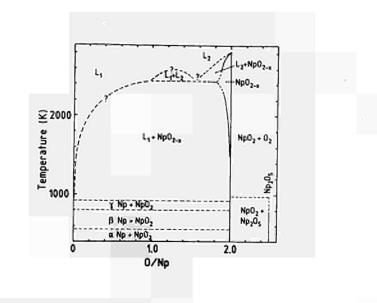


Fig. 3 Phase diagram of the system neptunium-oxygen

Some Recent Publications*

K.Richter, C.Sari, Phase Relationships in the Neptunium-Oxygen system, J.Nucl.Mater. 148(1987) 266

C.Sari, Grain Growth Kinetics in Uranium-Plutonium Mixed Oxides, J.Nucl.Mater. 137 (1986) 100

G.Pautasso, K.Richter, C.Sari, Investigation of the Reaction $UO_{2+x} + PuO_2 + C + N_2$ by Thermogravimetry, J.Nucl.Mater. (subm.)

* further relevant references may be found in "JRC Karlsruhe - List of Publications 1986/87" (EUR-Report 11790 EN, 1988)



Hj.Matzke

Materials Science

Competences

The Materials Science Section of the Transuranium Institute covers essentially six areas of research: irradiation testing, fuel rod modelling, microprobe analysis, electron microscopy, diffusion studies, and mechanical property studies.

The laboratory for irradiation testing specifies, designs, and fabricates irradiation devices and in-pile measuring equipment, organizes and supervises irradiation experiments, and evaluates the data obtained during these tests.

It collaborates closely with the team in charge of the mathematical modelling of fuel rod behaviour under normal, off-normal and accidental conditions for all reactor types, by paying special attention to the fission product behaviour (swelling, fission gas release) at high burn-up. Fully developed codes are available, also to external users.

The staff of the Institute's laboratories for microprobe analysis and electron microscopy is equipped and trained for handling radioactive (irradiated) specimens. Transmission electron microscopy can be performed with a resolution of better than 0.5 nm, and surface analysis by SEM with better than 6 nm. Techniques employed are lattice plane imaging and the use of replicas for surface studies.

Another group of investigations concernes the mechanical properties, in particular the deformation and fracture properties of radioactive materials.

The laboratory is specialised in the measurement of slow and fast diffusion processes in ceramics, including highly radioactive specimens, it employs highresolution alpha spectroscopy, SIMS, Rutherford backscattering, and other nuclear techniques.

The formation and the recovery upon annealing of irradiation damage in nuclear fuels and waste materials are studied.

Important Equipment

Shielded Electron Microprobe:

- Two electronprobe analysers; one shielded with heavy metal for the analysis of irradiated fuel (Fig. 1), the other enclosed in a glove box for the investigation of alpha-beta emitters;
- An energy dispersive analyser.

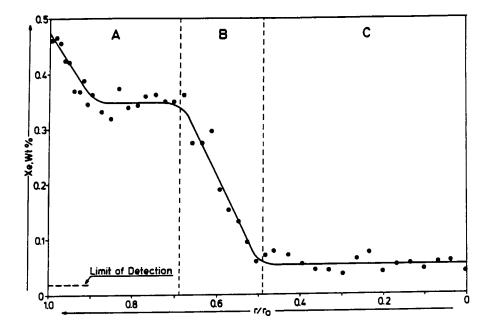


Fig. 1

Electron probe microanalysis: Radial distribution of xenon in a fuel pin after power transient

Diffusion Studies:

- High-resolution alpha-spectroscopy, SIMS, furnaces.

Electron Microscopy:

- Hitachi H700 HST 200kV STEM for the analysis of actinide-containing specimens and irradiated fuel samples (Fig. 2),
- Scanning electron microscope,
- Energy-dispersive analyser.

Mechanical Property Studies:

- Deformation apparatus up to 1600°C in a glove box,
- Indentation apparatus in a glove box,
- Short-rod fractometer.

Principal Achievements During the Last Three Years

Irradiation testing:

- Design of irradiation experiments CARLO, CARRO, POMPEI, NILOC1+2, SWELLAC
- Fabrication of ultrasonic temperature sensors for projects PAHR and SCARABEE (CEA)

Microprobe Analysis:

- Development of a technique for the analysis of grain boundary gas in irradiated nuclear fuel;
- Identification of the process (bubble interlinkage on grain boundaries) controlling gas release from high burn-up UO₂ under transient conditions.



Fig. 2

Hitachi H700HST electron microscope equipped for the investigation of irradiated fuel samples

Fuel Rod Modelling:

- Development of the codes TRANSURANUS, FUTURE, MITRA;
- Transfer of the TRANSURANUS code to licencing authorities and other users;
- Incorporation of the TRANSURANUS code into the European Accident Code (EAC).

Diffusion Studies:

- Studies of radiation-enhanced diffusion in nuclear fuels (Fig. 3)
- Measurements of actinide diffusion in waste glasses and repository salt.

Electron Microscopy:

- Detailed investigation of nucleation and growth of fission gas bubbles in oxide, carbide, and nitride fuel,
- Study of the precipitation of volatile fission products (I, Cs, Te),
- Investigation of the distribution of actinides in ceramic waste forms.

Mechanical Property Studies:

- Development of indentation techniques for fuel specimens and solidified radioactive waste.

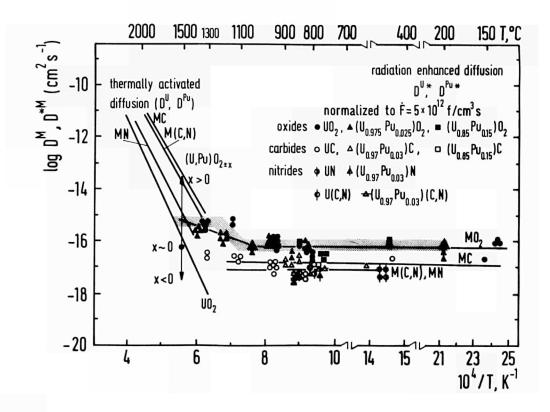


Fig. 3 Radiation enhanced diffusion in various nuclear fuels

Some Recent Publications*

Hj.Matzke, Science of Advanced LMFBR Fuels, A monograph on solid state physics, chemistry and technology of carbides, nitrides, and carbonitrides of uranium and plutonium, Elsevier Science Publishers, Amsterdam, 1986

P.T.Elton, K.Lassmann, Calculational Methods for Diffusional Gas Release, Nucl.Eng.Des.101 (1987) 259

Hj.Matzke, Nuclear Waste Materials, Chapt.12 in "Ion Beam Modification of Insulators", (G.Arnold and P.Mazzoldi, Eds.), Elsevier Science Publishers 1987

Hj.Matzke, Indentation Fracture and Mechanical Properties of Fuels and of Waste Ceramics and Glasses, European Applied Research Reports 7 (1987)

R.Cacciuffo, R.Coppola, F.Rustichelli, I.L.F.Ray, Investigation on He-Bubble Growth in Alpha-Implanted 1.4914 Steel, J.Nucl.Mater. (subm.)

C.T.Walker, P.Knappik, M.Mogensen, Concerning the Development of Grain Face Bubbles and Fission Gas Release in UO₂ Fuel, J.Nucl.Mater. (subm.)

Hj.Matzke, J.Ottaviani, D.Pellottiero, J.Ruault, Oxygen Potential of High Burn-Up Fast Breeder Oxide Fuel, J.Nucl.Mater. (subm.)

Hj.Matzke, Atomic Transport Properties in UO₂ and Mixed Oxides (U,Pu)O₂, J.Chem.Soc.,Faraday Trans. **83** 1987 (1121)

* further relevant references may be found in "JRC Karlsruhe - List of Publications 1986/87" (EUR-Report 11790 EN, 1988)



H.E.Schmidt

Applied Physics

Competences

The Applied Physics Section has two major tasks: To investigate and, if necessary, improve methods for the safe handling of radioactive, especially alphaactive, materials (with an accent on aerosol research), and to explore new areas of research beyond those defined by the specific research programmes, which fall in line with the overall mission of the Institute. The Group provides advice and technical support to other laboratories in the field of metrology and for the development of analytical methods. In addition, its capacities are available for work upon request from external customers.

In Aerosol Science, the Group is specialised in the investigation of aerosol formation (generation of test aerosols, release of radioactive aerosols from fires, resuspension of aerosols by mechanical forces), aerosol transport (in glove boxes and ventilated rooms), aerosol deposition (alpha recoil penetration of filters by radioactive aerosols, lung deposition and clearance, aerosol agglomeration), and the interaction gas/particle, always with special reference to nuclear fuel aerosols.

Present exploratory research deals with the acceleration of aerosol particle agglomeration in an acoustic field and the perspectives of this technique for air cleaning purposes.

The Group has expertise in laser technology, for example in the application of tunable lasers for analytical purposes (laser-induced mass spectrometry and molecular spectroscopy with CARS), and is contributing to the development of advanced optical pyrometers with ultrafast response.

The experimental work of the Group is complemented by extensive computer modelling, especially in the field of aerosol research.

Important Equipment

 A fully instrumented large scale (170 m³) fire facility is available for simulation experiments of aerosol dispersion during fires in nuclear facilities with non-radioactive aerosols) (Fig. 1);

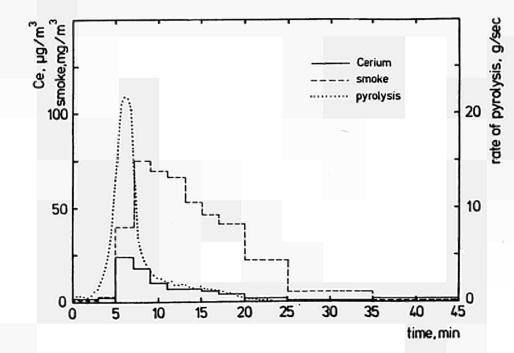


Fig. 1

Evolution of pyrolysis rate, cerium oxide aerosol concentration and smoke concentration as a function of time in a fire experiment, involving 4.9 kg of plexiglass. Ventilation rate 2000 m³/h (Ce-oxide was chosen as a stand-in for Pu-oxide)

- These experiments are complemented on a small scale in a glove box equipped for fire tests and dispersion studies with Plutonium-containing aerosols;
- Nuclear fuel aerosols of well defined characteristics for transport and dispersion studies are produced in specially developed generators,
- Optical, aerodynamic, diffusional, electrical, and other types of particle detectors are available for the determination of aerosol particle size distributions;
- Computer codes are used for investigating aerosol particle agglomeration processes and for predicting aerosol transport and particle deposition under various conditions;
- Acoustic aerosol agglomeration facilities employ high-power piezo-electric devices and pulsed chemical combustion engines for high-intensity sound generation.
- Advanced laser technology is used for improving molecular and mass-spectrometric analytical techniques.

Principal Achievements During the Last Three Years

- Measurement of resuspension of Pu particles from burning plexiglass;
- Interspecies comparison of lung clearance of radioactive particles;
- Investigation of lung clearance mechanisms for Pu-oxide particles in rats, in vivo and in vitro;

Calculation of flow field and aerosol distribution in a ventilated enclosure (glove box)(Fig. 2);

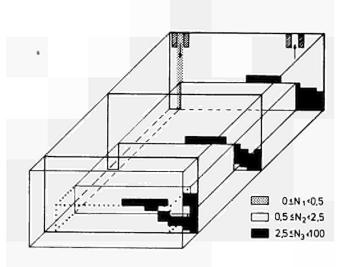
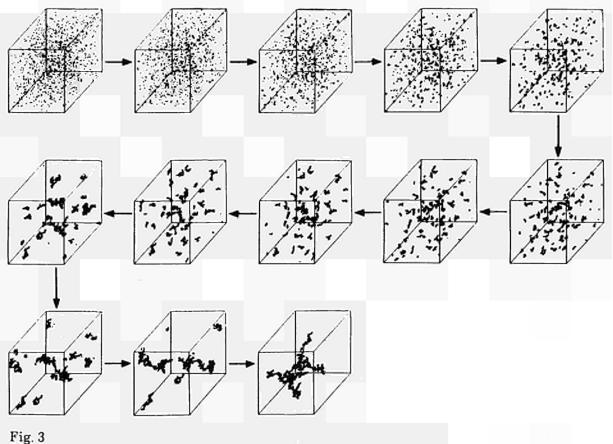


Fig. 2 Aerosol concentration in a ventilated enclosure (glove box) according to a 2-D computer model

- Generation of well defined nuclear fuel aerosols by comminution; Monte-Corlo modelling of aerosol agglomeration (Fig. 3);



3D-aerosol particle cluster-cluster agglomeration (model calculation)

- Clarification of mechanisms responsible for filter penetration of alpha-active aerosols;
- Systematic study of enhanced aerosol agglomeration in an acoustic field;
- Investigation of large-scale (radioactive) particle distribution in ventilated enclosures during fires;
- Study of (toxic) gas adsorption to aerosol particles and its enhancement by application of a sound field.

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S.Pickering, Particle Distributions in Simulated Agglomerations, J.Aerosol Sci. 18 (1987) 631

H.-L.Müller, A.Seidel, E.Drosselmeyer, G.Hotz, H.Thiele, S.Pickering, I.L.Ray, Zum biologischen Verhalten von Uran-Plutonium-Mischoxid Aerosolen: Inhalationsversuche mit Ratten und in vitro-Studien mit Alveolarmakrophagen, Report EUR 10846 DE (1987)

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S.Pickering, Resuspension of Uranium-Plutonium Oxide Particles from Burning Plexiglass, J.Aerosol Sci. 18, (1987) 927

J.F.Babelot, M.Hoch, The Spectral Emissivity of Metals and Non-Metals in the Wavelength Range 400 - 15000 Nanometers and their Total Emissivity, 11th European Conference on Thermophysical Properties, Umeå, Sweden 13 - 16 June 1988

* further relevant references may be found in "JRC Karlsruhe - List of Publications 1986/87" (EUR-Report 11790 EN, 1988)



M.Coquerelle

Hot Cell Laboratory

Capabilities

Sophisticated hot cell facilities and equipment for the examination of highly radioactive specimens are essential for R&D-programmes involving irradiated materials. The alpha-gamma hot cell laboratory of the European Institute for Transuranium Elements is one of the largest European facilities of its kind. It comprises a radiometallurgical and a radiochemical laboratory and is capable of performing destructive and non-destructive examinations of irradiated fuel pins and instrumented irradiation devices which have been irradiated in test facilities, as well as in commercial thermal reactors. In addition to nuclear fuel, the laboratory is in a position to examine cladding and structural materials, as well as waste forms (glasses and spent fuel). The facility can handle materials with a total gamma activity ranging from 10^4 to 10^6 Curie (at 1 MeV).

Important Equipment

In total 24 hot cells are arranged in three lines (Fig. 1). The a-tight stainless steel caissons of the radiometallurgical laboratory are constantly kept under high-purity nitrogen with less than 0.3% O₂ and less than 10 ppm of H₂O.

The radiometallurgy laboratory is equipped with two remotely controlled optical microscopes (Reichert), one of which is linked to a TAS (Leitz) image analyser (Fig. 2). The image analyser is mainly used for porosity determinations and for grain size measurements.

An alpha-gamma shielded scanning electron microscope (Jeol JSM 35 C) is available for the analysis of irradiated specimens with a maximum of 1 Curie (1 MeV) gamma activity. The instrument is equipped with an energy dispersive xray analyser (Tracor Northern) for identifying the constituents of micro-size phases.

The determination of fission gas release under both reducing and oxidizing atmosphere is possible by heating irradiated specimens to temperatures upto 2700°C in an induction furnace which is connected to a glove box with a gamma monitor and a quadrupole mass spectrometer.

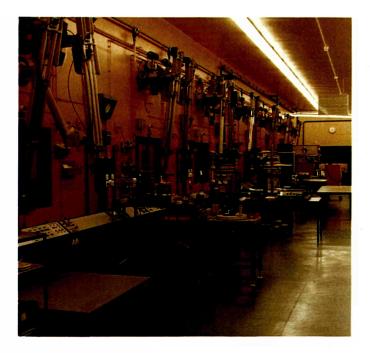


Fig. 1 View of the working area of the laboratory

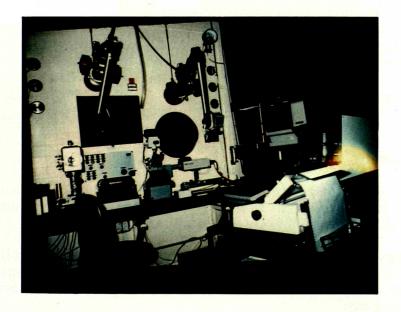


Fig. 2 Optical microscope with image analyser for irradiated specimens

Special equipment has been installed for the characterisation of waste glass and/or spent fuel. One cell with soxhlets is devoted to leach testing of waste forms (Fig. 3).

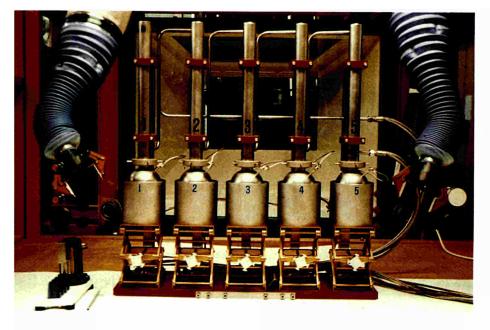


Fig 3 Facilities for performing leach tests on waste forms

Principal Achievements During the Last Three Years

- Investigation of the restructuring of mixed carbide and nitride fuels during irradiation;
- Analysis of fuel debris from the Three Mile Island reactor;
- Study of the fission gas behaviour in UO₂ irradiated to high burn-up (contract work for EPRI/BN) and under transient conditions;
- Characterisation of highly active glasses and performance of leach tests;
- Development of methods for dissolving mixed carbides and nitrides in HNO₃;
- Determination of micro structures and performance of radiochemical analyses on mixed minor actinide oxide fuels.

All technical capabilities and the major installations of the hot cell laboratory are summarized in the following table.

Technical capabilities of the alpha-gamma hot cell laboratory

Non-Destructive Examination of Fuel Pins and Instrumented Ca - visual inspection	apsules
- metrology - eddy current testing	
- x-radiography	
- gamma spectroscopy - gamma scanning.	
Annealing and Fission Product Release Tests - RF-induction and direct electrical heating furnaces	
- gamma flow counter - guadrupole mass spectrometer.	
- quadrupole mass spectrometer.	
Microstructure Examination of Fuels, Waste Forms, and Structure - optical microscopy	ural Materials
- image analysis	
 scanning electron microscopy (SEM) energy dispersive x-ray analysis (EDAX) 	
Isotopic and Chemical Analysis - alpha, beta, gamma spectroscopy	
- K-edge densitometry	
 mass spectrometry ICP spectrometry 	
 EMF measurement for oxygen potential determination O,C determination by IR spectroscopy 	
- N determination with a thermal conductivity cell	
Special Equipment	
- microsampling	
- laser cutting - soxhlets for leaching tests.	

Some Recent Publications*

M.Coquerelle, C.Ronchi, J.Sakellaridis, J.van de Laar, Analysis of the Fission Product Behaviour under Normal and Off-Normal Reactor Conditions, IAEA Report IGWTPT 27 (1987)

M.Coquerelle, C.Ronchi, Analysis of the Various Modes of Fission Product Release from LWR Fuel, Jahrestagung Kerntechnik '87, 2 - 4 June 1987, Karlsruhe, FRG

P.D.W.Bottomley, M.Coquerelle, Metallurgical Examination of Bore Samples from the TMI-2 Reactor Core, ANS/ENS Topical Meeting on the Three Mile Island Accident, 30 October - 4 November 1987, Washington DC, USA

* further relevant references may be found in "JRC Karlsruhe - List of Publications 1986/87" (EUR-Report 11790 EN, 1988)



K.Richter

Nuclear Technology

Competences

An important asset for an Institute engaged in research and development on nuclear fuels is the availability of well characterized specimens for the various investigations. These specimens must satisfy the requirements of scientists performing basic studies as well as those of the engineer who investigates the irradiation performance of various fuel types.

The Nuclear Technology Service of the Institute has a long-standing experience in the preparation of oxide, carbide, and nitride fuels on the basis of uranium and/or plutonium, determined for irradiation experiments in fuel pins or specially designed capsules, or for the study of out-of-pile materials properties. It is equipped for the characterisation and for quality control of ceramic powders, fuels and fuel pins and is permanently engaged in the further refinement of established fabrication procedures, the development of new fuel materials according to increased safety standards, and the detailed investigation of the physics and the chemistry of the various process steps.

Besides this, the Group is charged with the reprocessing of the Institute's nonirradiated solid and liquid laboratory waste, which implies the recovery of fissile materials and the conditioning of the waste for final storage.

Special activities of the Group deal with the preparation of Cf-sources, the encapsulation of highly active materials (Am), the fabrication of standards and sources for destructive and non-destructive measurements, and the preparation of metallic actinide-containing samples.

Important Equipment

3 hydraulic presses (20 - 60 to) (Fig. 1);

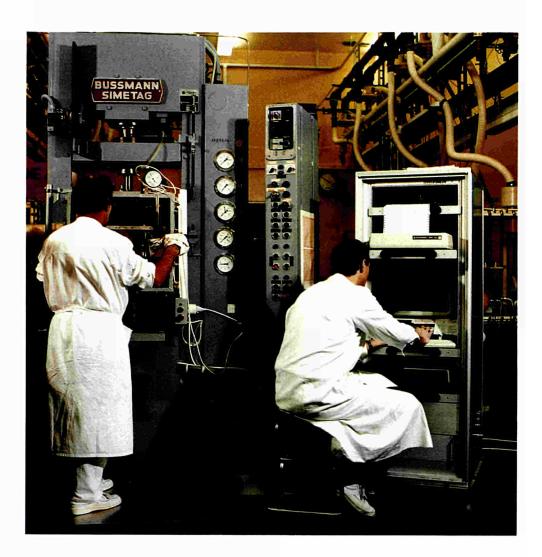


Fig. 1

Hydraulic press for compaction of (U,Pu) fuel pellets

- 3 vacuum sintering furnaces for temperatures up to 1800°C (load 1 kg);
- 3 ceramic-lined furnaces with Mo-heating elements for temperatures up to 1700 °C (load 2 kg);
- 1 induction furnace (load 2 kg); 2 electron beam welding installations (Fig. 2);
- 2 TIG (tungsten inert gas) -He arc welding machines;
- 1 sol-gel dropping installation; -
- 1 cell for the encapsulation of highly active material (Cf, Cm);
- 2 ion exchange installations for Pu purification and separation;
- various equipment for the characterisation and the quality control of powders, unirradiated fuel materials and fuel pins in altogether 10 laboratories. -



Fig. 2 Installation for welding of fuel pin end caps

Principal Achievements During the Last Three Years

- Development of a gel-supported precipitation process (in collaboration with ENEA-AGIP) for $(U,Pu)O_2$ and $(U,Am)O_2$ - $(U,Am,Np)O_2$ fuels;
- Development of a fabrication process for (U,Pu)-nitrides with low oxygen and
- carbon content (NILOC); Development of a new "direct pressing" method for nitrides and carbides (eliminating one of the process steps of the "classical" procedure) (Fig.3); Fabrication of test pins,for the following irradiation experiments: CARLO, CARRO, KNKII (19 pin carbide bundle), NILOC 1 and 2, NIMPHE 1 and 2, SUPERFACT (two versions).

CARBIDES - NITRIDES (U,Pu)C (U,Pu)N

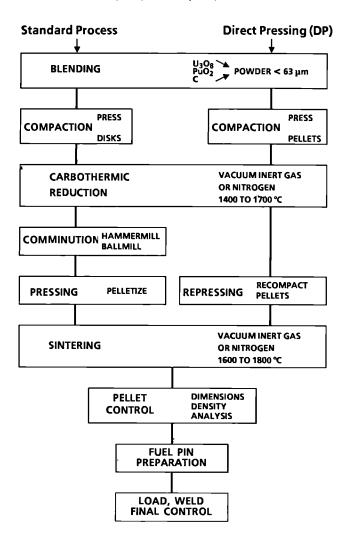


Fig. 3

Flow sheets, comparing the "direct pressing" procedure for the preparation of advanced fuel with the conventional technique

Some Recent Publications*

K.Richter, H.Blank, Fast Reactor Fuel Fabrication Processes and Development, Nuclear Europe 3(1987) K.Richter, U.Benedict, J.F.Gueugnon, H.Kutter, C.Sari, G.Mühling, Specifi-

cation, Characterisation, and Fabrication of (U,Pu)C Fuel, J.Nucl.Mater. 153 (1988) 205

H.Blank, K.Richter, New Concepts for Fast Reactor Fuels, Jahrestagung Kerntechnik `88, 17 - 19 May 1988, Travemünde, FRG

* further relevant references may be found in "JRC Karlsruhe - List of Publications 1986/87" (EUR-Report 11790 EN, 1988)

Some Figures and Facts about the Institute (Status October 1988)

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1. The Staff

Total: 200

by activities

 * Thermophysics: 15 * Fuel Characterisation: 5 * Materials Science: 13 * Applied Physics: 7 * Analyt. & Radiochem.: 20 * Actinide Research: 12 	07
* Applied Physics: 7 * Analyt. & Radiochem.: 20	
* Actinide Research: 12	
* High Pressure Studies: 3	
* Chemical Technology: 9	
* Hot Cells: 10	
* Nuclear Technology: 13	

by training

-	university trained:	44
-	technicians, administrators:	153
-	skilled workers:	3

by nationality (in %)

-	В	:	11.2
-	D	:	45.5
-	DK	:	0.5
-	\mathbf{E}	:	1.6
-	\mathbf{F}	:	16.0
-	GB	:	4.8
-	GR	:	0.5
-	I	:	8.6
-	IRL	:	0.5
-	LUX	:	3.8
-	\mathbf{NL}	:	7.0

2. The Laboratory

2.1 Buildings

Total surface: $16\ 000\ m^2$ Total volume: $188\ 000\ m^3$

2.2 Laboratories

Number of alpha-laboratories:	30
number of "cold" laboratories in the controlled area:	16
number of hot cells:	14
number of installed boxes (provided with gloves	
or manipulators; 6 of them shielded with lead or water)	420

4. Installations and Consumption

4.1 Nitrogen purification plants

total throughput	600 000	m³/h
* hot cells:	2000	m ³ /h
* glove boxes:	900	m³/h

4.2 Consumption

Electrical power:	11	GWh/a
Heating power :	71000	GJ/a

5. The Radiation Protection Service

Hand and foot monitors in continuos operation	42
Continuous air monitoring posts	42
Dust collecting filters measured per year	20000
Dosimeters checked per year	10000
Various specimens analysed for radiation	
per year	3500
Smear tests evaluated per year	9000
Area of laboratory floor under surveillance	150000 m^2
Signals (mostly tests) arriving	
at the central monitoring desk per year	10000
Instructions on radiation protection matters	
given per year	250

6. Publications

	1983	1984	1985	1986	1987
Contributions to journals	41	39	29	30	34
Books and monographs	2	4	10	3	5
Contributions to conferences	54	45	69	70	56
Various reports	19	15	9	21	19

7. Collaborations with External Organisations

- 7.1 Belgium
 - Belgonucléaire, Brussels
 - CEŇ Mol
 - University of Antwerp
 - University of Liège
 - University of Namur
- 7.2 Canada
 - Atomic Energy of Canada Ltd
- 7.3 Czechoslovakia
 - University of Prague
- 7.4 Denmark

 - Nordita, CopenhagenRisø National Laboratory
 - Technical University of Lingby
 - University of Copenhagen
- 7.5 Federal Republic of Germany Battelle Institut, Frankfurt
 - - KfA Jülich
 - *Institut für Festkörperforschung
 - *Institut für Chemische Technologie
 - KfK Karlsruhe
 - *Institut für Heisse Chemie
 - *Institut für Material- und Festkörperforschung
 - *Institut für Nukleare Entsorgungstechnik
 - *Institut für Neutronenphysik und Reaktortechnik
 - *Institut für Genetik und Toxikologie
 - *Institut für Nukleare Festkörperphysik
 - *Institut für Radiochemie

*Projekt Schneller Brüter

- Max-Planck-Institut für Chemie, Mainz
- Max-Planck-Institut für Festkörperphysik, Stuttgart
- Max-Planck-Institut für Metallforschung, Stuttgart
- Siemens-KWU, Erlangen
- Technische Universität München
- TÜV BadenTÜV Bayern
- TÜV Hamburg
- Freie Universität Berlin
- Universität Erlangen
- Universität Karlsruhe *Physikalisches Institut *Institut für Chemische Technik
- Universität Paderborn
- Universität Stuttgart
- 7.6 France
 - CEA, CEN Cadarache
 - CEA, CEN Grenoble
 - CEA, CEN Saclay
 - CNRS Strasbourg
 - CNRS Orsay
 - CNRS Villetaneuse
 - ILL Grenoble
 - Institut Curie, Paris
 - LURE, Orsay
 - Péchiney-CÓMHORHEX
 - University of Strasbourg
- 7.7 Greece
 - University of Patras
- 7.8 Italy
 - Ente Nazionale Energia Alternative, Casaccia
 - Istituto Nazionale Fisica Nucleare, Frascati
 - LAMEL, Bologna
 - Lavoro e Ambiente, Bologna
 - University of Bologna
 - Politecnico Milano
 - University of Padova
 - University of Parma
- 7.9 Japan
 - CRIEPI
 - National Laboratory of Metrology
- 7.10 Netherlands
 - Philips, Eindhoven
 - University of Amsterdam

- 7.11 Portugal - ICEN-LNTI, Lisboa
- 7.12 Spain - Instituto de Acustica, CSIC, Madrid
- 7.13 Switzerland
 - ETH Zürich
 - University of Geneva
- 7.14 United Kingdom
 - AERE Harwell
 - Birbeck College
 - Rutherford-Appleton Laboratory, Oxford
 University of Birmingham
 University of Salford

 - University of Warwick, Coventry

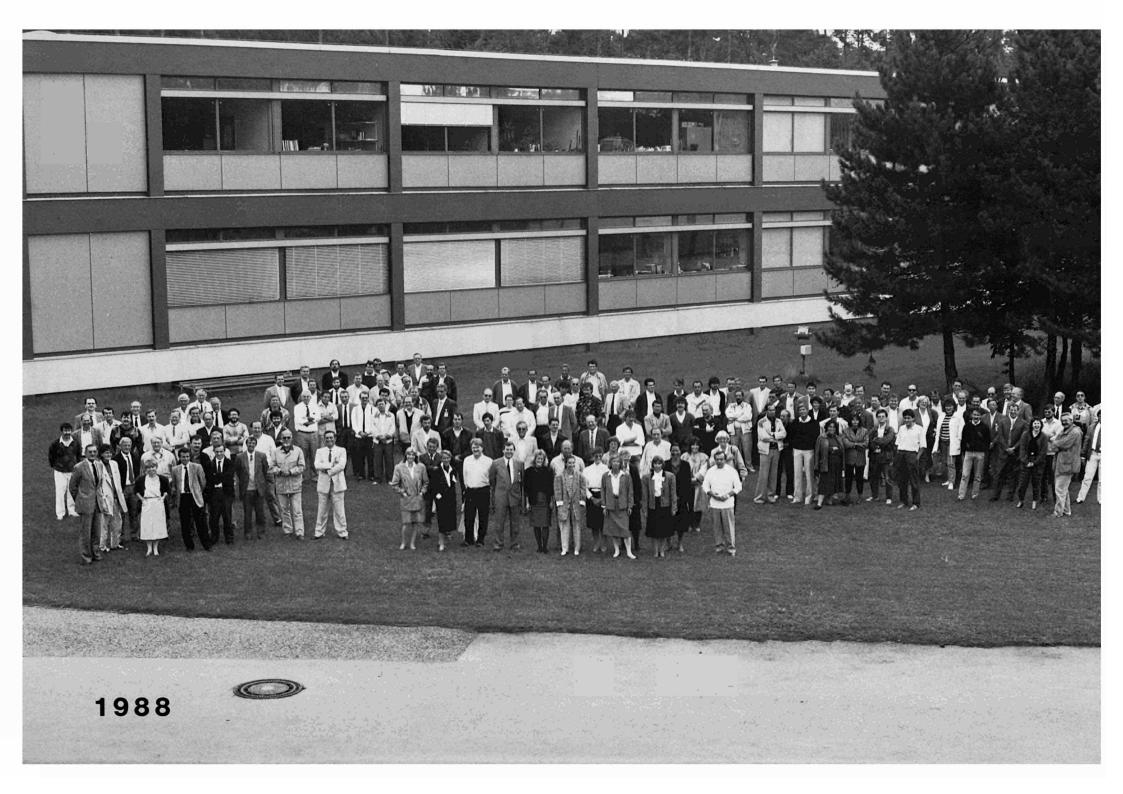
- 7.15 United States of America Argonne National Laboratory Battelle Columbus

 - EG&G, Idaho Falls
 - Lawrence Livermore National Laboratory
 - Los Alamos National Laboratory
 - Oak Ridge National Laboratory
 - University of Cincinnati

7.16 International Organisations

- CEC, DCS Luxembourg
- JRC Ispra
- JRC Petten
- JRC Geel
- IAEA Vienna
- OECD, Paris





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