EURATOM

INSTITUTE FOR TRANSURANIUM ELEMENTS KARLSRUHE

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Aerial photograph of the Karlsruhe Nuclear Research Centre with the European Institute for Transuranium Elements in the foreground, lower left.
Cover: The T[illegible] 1975

THE EUROPEAN INSTITUTE

FOR TRANSURANIUM ELEMENTS

(E U R A T O M)

KARLSRUHE

1975

How to get to the Institute

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CONTENTS

Published by the Commission of the European Communities
Directorate-General Scientific and Technical Information
and Information Management
Luxembourg, 1975

Compiled and edited by H.E. Schmidt

A research laboratory financed by public means should give from time to time a compre hensive description of its activities.

The staff of the European Institute for Transuranium Elements, which is the Karlsruhe establishment of the Joint Nuclear Research Centre of the European Community EURATOM, has, therefore, compiled this booklet for the information of the interested public in general, as well as for colleagues, visitors and friends.

The first part describes the objectives of the Institute, its historical and geographical background, its structure, and its operation. In the second part, a more detailed account of the activities of the Institute is given for the interested specialist.

The mission of the European Institute for Transuranium Elements is twofold:

1. Much of the work is concentrated on plutonium and its compounds, especially on those which are considered for use in nuclear reactors, mainly fast neutron breeders. Although the laboratory has established its competence for industrial fuel element production by the fabrication of the fuel elements for the French MASURCA reactor, it is not in competition with industry ;rather it engages in medium and long-term research programmes dealing mainly with advanced nuclear fuels, such as carbides and nitrides.

2. The Laboratory maintains a general competence in the field of transuranium elements. Chemical operations and physical measurements are carried out on heavy elements and their compounds up to californium.

The Laboratory performs its tasks in close cooperation with national laboratories. Its activity is coordinated, and kept useful and complementary to national research programmes by a Programme Advisory Committee established by the Council of Ministers of the European Communities . The committee is staffed with representatives from science, industry and government of the member states.

The European Institute for Transuranium Elements in Karlsruhe constitutes an example of sucessful European Community cooperation in the field of nuclear research. Since there exist only a few laboratories of this type in Europe (their construction and operation being time consuming and costly), it can be assumed that the Institute will keep its place within the scientific and technical community for many years to come.

Karlsruhe, in April 1975

R.Lindner

I. The Institute, its Background and How it Works

The Transuranium Elements

Before 1940, the Periodic Table of the chemical elements ended with uranium, the heaviest element to have survived since the origin of the solar system. After the discovery of neutrons (1932) there were numerous unsuccessful attempts to synthesize elements beyond uranium. The search for transuranium elements led to the discovery of nuclear fission by HAHN and STRASSMANN in 1939. As a result of experiments to measure the energy liberated during the fission of uranium nuclei by neutrons, McMILLAN observed the formation of radionuclides whose chemical properties did not agree with those of any natural element. In cooperation with G.T. SEABORG, these artificial elements were identified as TRANSURANIUM ELEMENTS. In analogy to the sequence of the planets, the first transuranium elements were called neptunium (Np) and plutonium (Pu). In 1951, McMILLAN and SEABORG were awarded the Nobel prize in recognition of their discovery .

Formation of transuranium elements

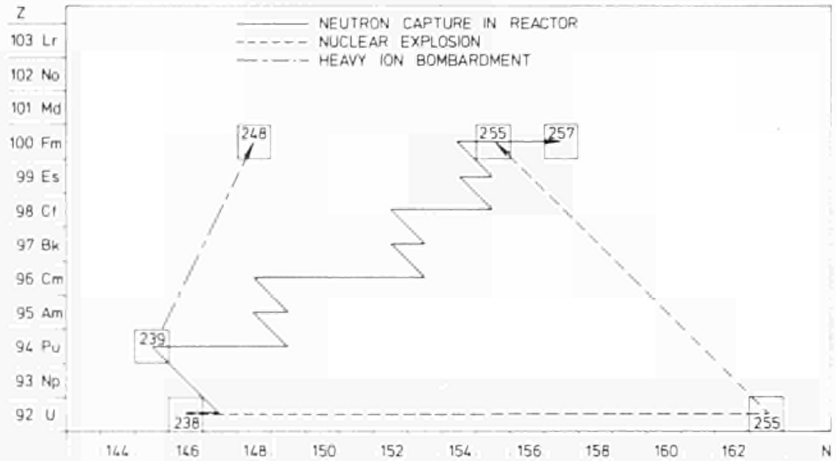
A chemical element is characterized by its atomic number Z, i.e., the number of protons in the nucleus. The atomic mass is determined by the sum of the protons (Z) and neutrons (N) in the nucleus. To synthesize transuranium elements, proton numbers greater than 92 have to be attained by nuclear reactions.

The most important nuclear reactions used to build up transuranium elements are:
a) stepwise neutron capture and decay of β-instable, heavy nuclides during irradiation in a nuclear reactor,
b) multiple neutron capture during nuclear explosions or during rapid nucleosynthesis in stars.
c) bombardment with heavy nuclei in particle accelerators.

Most of the first transuranium nuclides were synthesized by the nuclear reaction(a):

$$^{238}_{92}U + {}^{1}_{0}n = {}^{239}_{92}U \xrightarrow{\beta^-} {}^{239}_{93}Np \xrightarrow{\beta^-} {}^{239}_{94}Pu$$

This reaction results in the formation of fissile plutonium in nuclear fuel, which increases the lifetime of fuel elements or, in breeder reactors, generates more fissile material in the blanket than is consumed in the core.



Mechanisms of formation of transuranium nuclides (Z: Atomic number, N: neutron number)

Elements 99 (einsteinium, Es) and 100 (fermium, Fm) were discovered in the debris of the first thermonuclear explosion; they had been formed according to reaction (b) :

$$^{238}_{92}U + 15\,^{1}_{0}n = ^{253}_{92}U \xrightarrow{8\,\beta^-} ^{253}_{100}Fm$$

Elements with atomic numbers higher than 100 can only be synthesized with accelerated particles (reaction c):

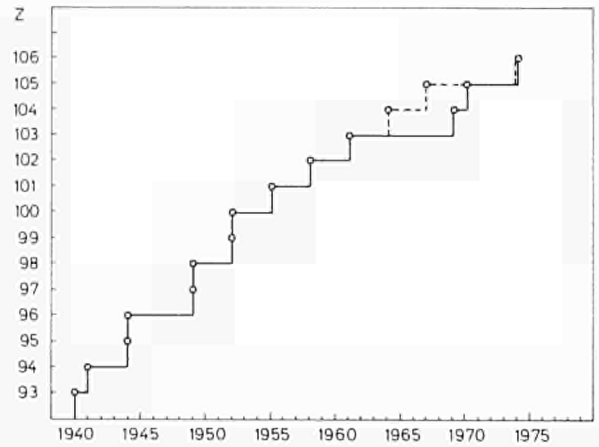
$$^{244}_{94}Pu + ^{16}_{8}O = ^{255}_{102}No + 5\,^{1}_{0}n$$

Here a plutonium target is bombarded with oxygen to produce element 102, nobelium.

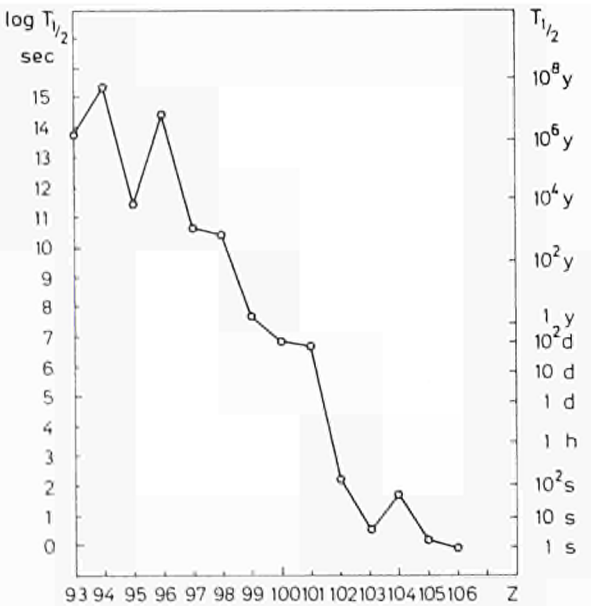
For several years, groups working at Berkeley (USA) and at Dubna (USSR) have both claimed priority for the synthesis of the latest transuranium elements to be found. In 1974, both groups independently reported the discovery of element 106.

Nuclear Stability and Superelements

Up to element 106, the half lives of the longest-lived nuclide of each of the transuranium elements decrease with increasing Z, from more than a million years for the most stable



History of the discovery of heavy elements (dotted lines indicate soviet work)



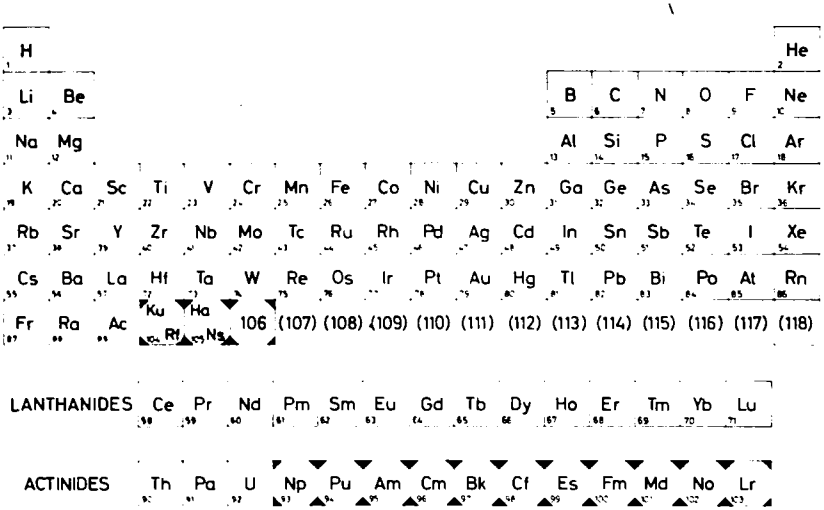
Variation of half-lives of the longest-lived transuranium nuclides with atomic number

nuclide of neptunium down to less than a year for einsteinium. Heavier transuranium elements have a fleeting existence measurable only in fractions of a second. But recent calculations (based on the nuclear shell model and the existence of magic numbers) suggest that elements with atomic numbers around 110 - 114 might be more stable again. So far, however, attempts to synthezise or to detect in nature these superelements on what SEABORG calls **islands of relative stability** in a **sea of unstability** have failed.

The Actinide Concept of Transuranium Elements

Before their discovery, it was assumed that the transuranium elements would be homologues of the elements Re, Os, Ir in groups VII and VIII of the Periodic Table. This classification was put in doubt when the newly synthesized elements proved to be similar to the Rare Earth elements. The chemical similarity of the Rare Earths results from the similarity of the configuration of the outer electrons; the 14 Rare Earth elements following lanthanum differ only by the number of electrons in the inner 4f shell. The **lanthanides**, therefore, are 4f elements. By analogy, SEABORG suggested that the transuranium elements belong to a family of 5f elements. The 14 elements following Ac (actinium, $Z=89$) in the Periodic Table (up to $Z=103$) are characterized by increasing occupation of the 5f shell and are called **actinides**.

SEABORG's actinide concept was very successful; it allowed the newly synthesized elements to be fitted into the Periodic Table in an apparently logical way, it helped to find methods to synthesize and identify heavier elements, and it found its ultimate confirmation in the correct prediction of the chemical properties of element 104, which turned out to be the homologue of Hf. Hf is the first element after the lanthanides in the Periodic Table, and therefore element 104 is the first transactinide.

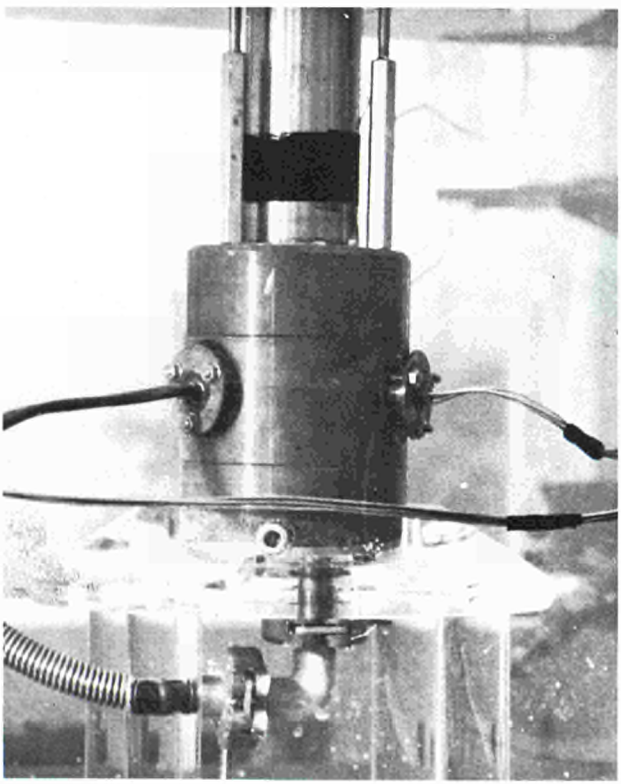


Position of the actinides and transactinides in the Periodic Table

Despite its success, the actinide concept in its initial form was contested. With increasing knowledge of the chemical properties, fundamental differences, not only between lanthanides and actinides, but also between light and heavy actinides became evident. Whereas most of the lanthanide ions exhibit the oxidation number three, some of the early members of the actinide family can be oxidized to the heptavalent state. On the other hand, there is evidence for the existence of monovalent mendelevium ($Z=101$). Furthermore, the lanthanide metals crystallize predominantly in close packed structures; although these structures can be observed with transplutonium metals (Am, Cm), the lighter actinides (Np, Pu) possess unusual crystal structures of low geometry. Although both lanthanides and actinides are f-elements, there are differences determined by fundamental differences between 4f and 5f electrons.

Application of Transuranium Elements

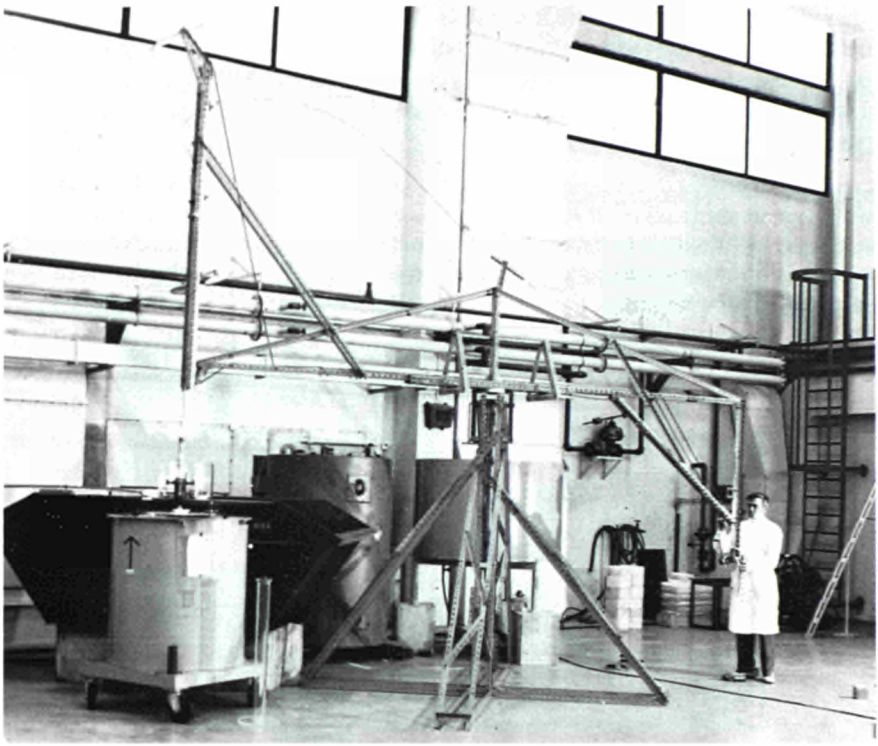
The practical application of transuranium elements depends on their nuclear properties. The nuclei of transuranium elements are unstable, some of them can be fissioned by the absorption of slow neutrons and all decay spontaneously, i.e. they are radioactive.



Radionuclide battery fuelled with ^{242}Cm-oxide

The fissibility of ^{239}Pu permits the best use of natural uranium re sources for nuclear energy production. The conversion of non fissile ^{238}U to ^{239}Pu will help to meet future energy demands in three different ways:

- It can be burnt in situ (i.e. in uranium based fuel elements where it is formed by nuclear reactions) and thus helps to increase the lifetime of the fuel element;



Spontaneous fission neutron irradiation facility

- it can replace ^{235}U in enriched fuel; this is the recycling of plutonium recovered from spent fuel elements;
- it can be burnt in the core of fast breeder reactors; by this means, more fissile material can be formed in the natural uranium blanket around the core than is burnt inside the core.

The radioactivity of transuranium elements is characterized by α-decay and spontaneous fission, accompanied by x-ray and gamma radiation. The α-decay energy of ^{238}Pu is being used as a power source in space technology and medicine. Gamma (^{241}Am) and neutron (^{252}Cf) sources are increasingly being applied in analysis, medicine and technology.

Present Problems in Transuranium Element Research

Transuranium elements have been discovered by nuclear physicists and radiochemists. In the meantime, theoretical chemists, solid state physicists, and chemical and nuclear engineers have become interested in the fascinating properties of these elements and in the possibilities for their application.

Plutonium-bearing fuels for future reactors are being developed and tested. Mixed uranium-plutonium oxide fuel is already used in prototype breeder reactors, the economy of which still has to be improved. The second generation of breeder fuel will contain mixed carbides or nitrides. Such advanced fuel is expected to be superior to oxide fuel because of its better breeding ratio and lower in-pile inventory, important factors in view of rapidly increasing energy demands.

Various physical and chemical methods of investigation are being combined to gain a better understanding of the chemical bonding in transuranium elements and their compounds. New concepts for the final disposal of long-lived radioactive transuranium isotopes that accumulate in nuclear waste are being discussed.

The European Institute for Transuranium Elements is contributing to the solution of many of these problems.

The European Communities and their Treaties

The treaties for the foundation of the European Economic Community and the European Atomic Energy Community were signed in Rome on March 25, 1957 by the six original member states, i.e. Belgium, Germany, France, Italy, Luxembourg and The Netherlands (these states had already founded the European Coal and Steel Community in 1951).

Signature of the Rome Treaties, March 25,1957

On April 8, 1965, the member states agreed that the three Communities should have one common Council of Ministers and one common Commission of the European Communities, which started its work on January 1, 1968. On January 22, 1972, representatives from the six original member states and from Denmark, Norway, Ireland and the United Kingdom decided that the four latter states should become full members of all three Communities, the agreement coming into effect after deposition of the ratification documents on January 1, 1973. This was indeed the case, with the exception of Norway.

The European Atomic Energy Community Treaty

Its preamble states that the Treaty was concluded

- taking into consideration that nuclear energy is an indispensable source for the development and the vitalization of the economy and for progress in peace
- with the determination to create the most favourable conditions for the development of a powerful industry, which extends the energy production, modernises technology and contributes to the welfare of the nations...

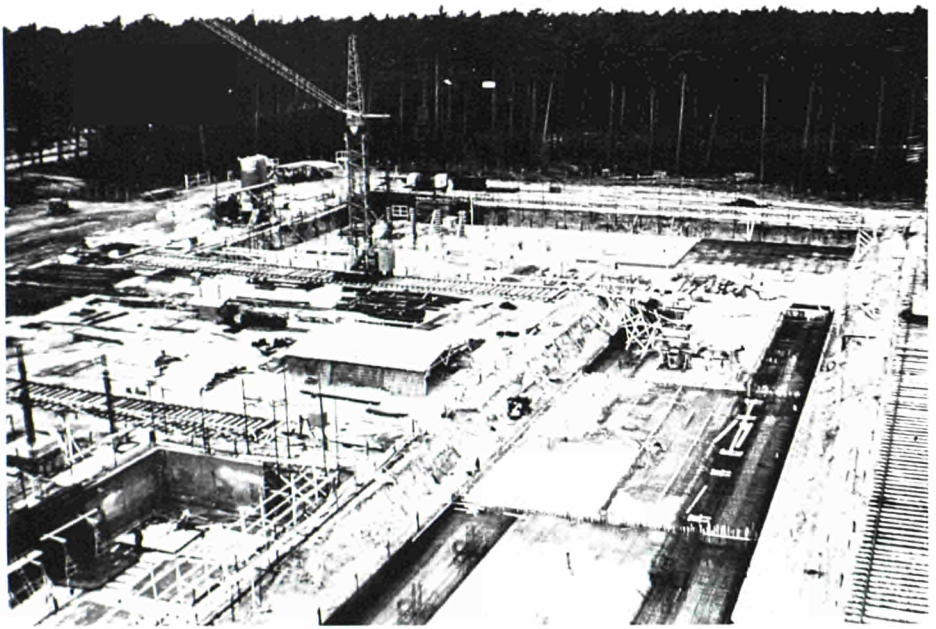
Article 1 defines the objectives of the Community: ... to contribute to an increase in the standard of living in the member states and to the development of the relations to other countries by preparing the grounds for a rapid development of nuclear industries.
Furthermore, Article 2 reads that in fulfilment of its duties, defined in the Treaty, the Community has to encourage research and assure the dissemination of technological knowledge.
According to Article 4, the Commission has to stipulate and facilitate nuclear research in the member states and to complement these efforts by its own research and training programme.
Article 8 provides for the creation of a **Joint Research Centre** which executes the research programmes of the Community and other tasks defined by the Commission.

In Appendix I of the Treaty, the areas of research according to Article 4 are specified; in Appendix IV reference is made to the treatment of radioactive materials, and under items (2) and (3) the **chemistry and metallurgy of plutonium** and the procedures for the **preparation and the chemistry of other transuranium elements** are mentioned.

This is the basis for the creation and the operation of the European Institute for Transuranium Elements.

Some Historic Dates and Facts

On December 21, 1960, a treaty was signed between the government of the German Federal Republic and the Euratom Commission concerning the construction and operation of a transuranium laboratory on the site of the German Nuclear Research Centre near Karlsruhe. The ground and the building were to be provided by the national counterpart, with the construction work being supervised and coordinated by a mixed planning group, composed of delegates from the Euratom Commission and the Gesellschaft fuer Kernforschung; the laboratory was to be operated by the Commission.
The mixed planning group began its work early in 1961. The ground was broken and



The TU under construction 1963

the cornerstone laid on April 1, 1963. Plutonium was first introduced into one of the glove-boxes of the Laboratory on February 10, 1965.
The hot cells became operative in 1966. All construction work was finished in the course of 1967.

The first irradiated fuel rod is delivered to the Institute (1966)

The first Director of the laboratory was the French physicist Jean Blin[1].

In the early sixties, during the period of construction, many of the future staff members worked as guest scientists in the USA (Hanford, Argonne, Berkeley), France (Saclay,

One of the first visitors: Glenn T. Seaborg (on the right, together with Dr. Blin)

Fontenay-aux-Roses, Cadarache), Belgium (Mol and Liege) and in the United Kingdom (Harwell).

The first significant operation carried out at the Institute to test and demonstrate its technological competence and capacity was the fabrication of 2100 fuel rods for the French zero power reactor MASURCA in 1967.

The Institute, Facts and Figures

Personnel

In 1974, the staff of the laboratory comprised 210 persons of eight different nationalities, including 43 university-trained engineers and scientists.

The evolution of the distribution of nationalities among the members of the Institute is shown in the following table.

[1] now Head of the Metallurgy Department at the Centre d'Etudes Nucléaires de Grenoble, France

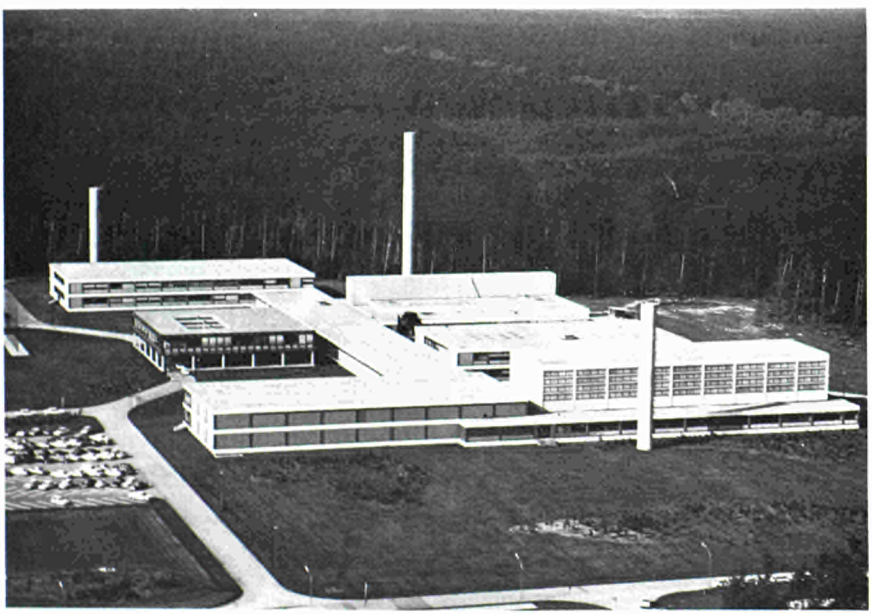
	B	D	DK	F	GB	I	L	NL
1966	12	29	-	19	-	17	3	20
1968	16	27	-	21	1	15	3	17
1970	16	29	-	19	5	14	3	14
1972	16	29	-	20	4	15	3	13
1974	13	25	1	17	8	22	4	10

Distribution of the personnel among nationalities (including students and guest scientists, not including local employees) in %

Between 1970 and 1975 more than 80 student trainees and guest scientists from 10 different countries spent periods between three months and two years working at the Institute.

Buildings

The buildings cover a surface of 16 000 m^2. The total volume is 188 000 m^3. The laboratory is divided into seven wings each with well defined specifications:

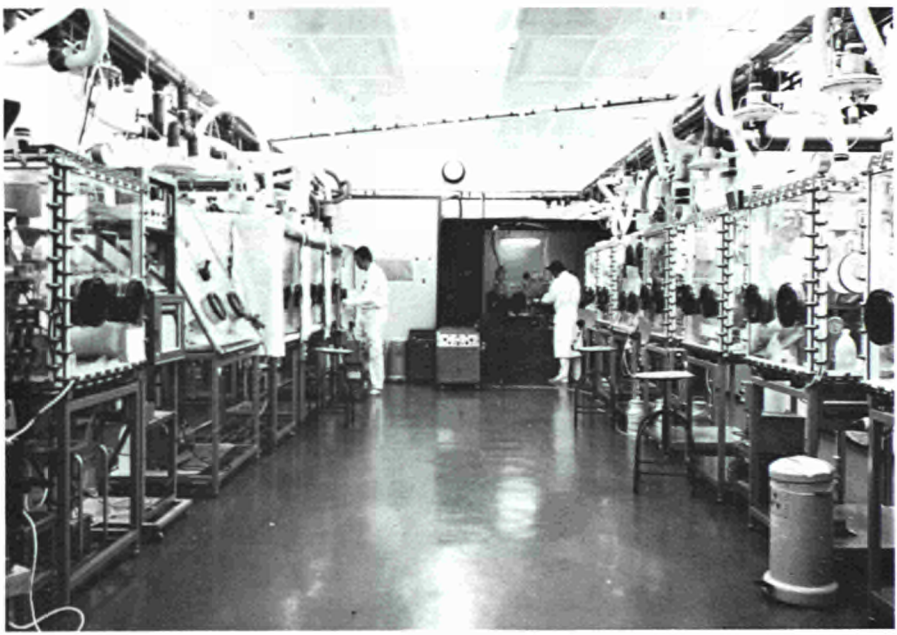


The European Institute for Transuranium Elements, as it is today

Three wings are provided for the manipulation of non-irradiated plutonium and transplutonium elements, one wing for technological operations, and a fifth wing is equipped with hot cells for handling highly radioactive materials. The administration and an auditorium for 70 persons are located outside the controlled zone.

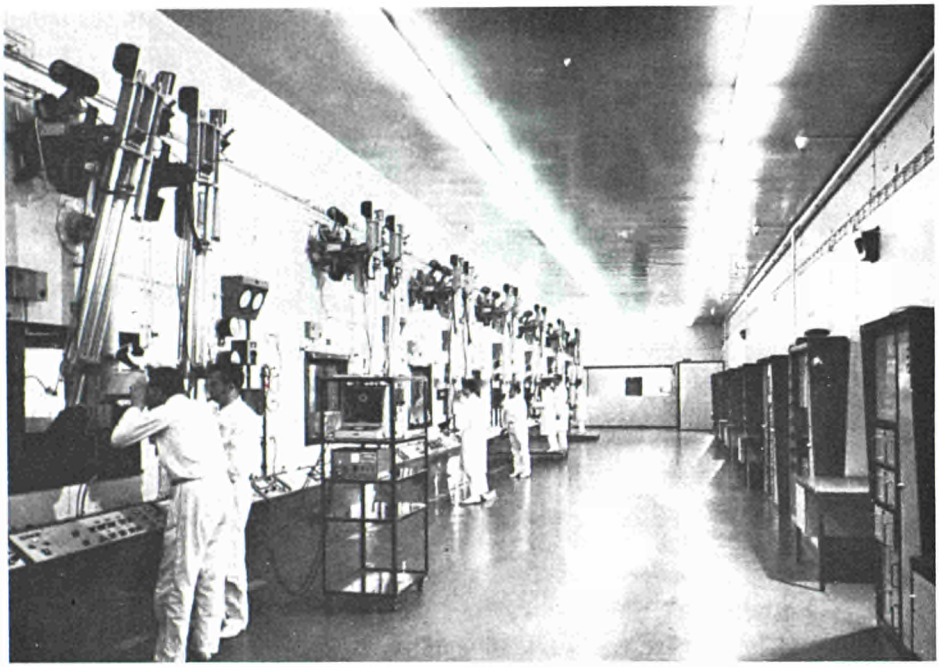
Laboratories

The Institute has 25 α-laboratories with an effective total surface of 2540 m^2. Additional hot laboratory space of 330 m^2 is provided by the caissons (α-tight steel structures which house groups of glove boxes) installed in the technology hall in wing G.



One of the Institute's α-Laboratories

The α-laboratories and the caissons are equipped with a total of about 600 glove-boxes, 40 % of which contain an inert gas atmosphere (argon or nitrogen).
Theαβγ-laboratory has 20 hot cells. Their principal characteristics and their designation are listed below.



The Hot Cell Laboratory, north-row

All cells, except for the disassembly cell, are α,β,γ-tight.
In chain II, biological protection is provided for the handling of transplutonium elements

	Biological protection Ci (γ) at 0.8 MeV	Number of Cells	Total working surface (m^2)	Atmosphere	Principal use
Chain I	10^6	7	75	nitrogen (except for the disassembly cell)	Disassembly, non-destructive examination, metallography, microscopy, machining
Chain II	10^4	6	23	air	Dissolution and separation for chemical analyses, separation of transplutonium elements
Chain III	10^2	5	11	nitrogen	metallography, microscopy, special examinations
Chain IV	10	2	4	nitrogen	decontamination and preparation of microprobe samples

Hot cell specifications

Man at work on alpha-tight glove-boxes

in quantities equivalent to 5 mg of ^{252}Cf. Another cell for this purpose with two operating platforms is installed in the technology hall; its biological protection consists of 60 cm of water.

For the handling of transplutonium elements with lower γ-activity, a line of special glove boxes is equipped with master-slave manipulators; the biological protection is provided by 25 cm of water.

Technical Installations

Two design features of the laboratory are especially noteworthy:

- Seven ventilation installations (one per wing) provide for increasing air pressure differences between non-active zones, active laboratories, glove boxes and hot cells, with a total air flow of 600 000 m^3/h.
- Two nitrogen purification plants are in operation. One of them with a capacity of 2000 m^3/h is coupled to the α-tight caissons of the hot cell laboratory, the other delivers up to 1200 m^3/h of pure nitrogen to about 250 gloveboxes. Gas which has been processed in these plants contains 0,5 % of oxygen and only a few ppm of water vapour.

Cost

The **construction** costs for the laboratory amounted to 16 million UC in 1967 [+]. This sum takes into account all buildings and all fixed technical installations, but not the mobile laboratory equipment.

The cost for **maintenance and operation** of the laboratory was 1.2 million UC in 1974.

Some Figures (for the year 1974)

- electrical power consumption	8 757 780 kWh
- room heating	54 858 GJ
- nitrogen consumption	151 872 m^3
- hydrogen consumption	34 580 m^3

[+] 1 UC (unit of account) = 3.66 DM (1966)

In this and other contexts the Laboratory is indebted to the German Nuclear Research Centre of the Gesellschaft fuer Kernforschung with which very good relations are maintained.

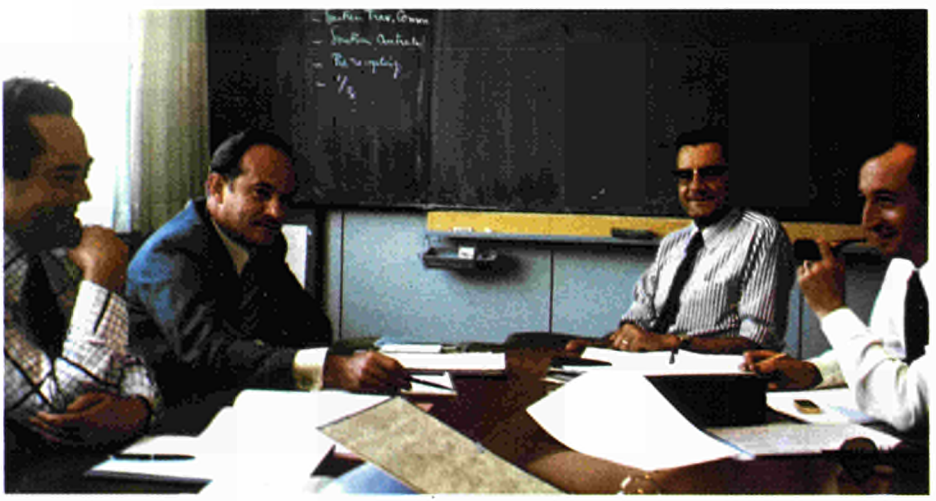
Management of the Plutonium and Transplutonium Programme

The Director of the Institute for Transuranium Elements
- responds to the needs and suggestions of a number of authorities and groups and
- controls at the same time the internal structure of the laboratory in order to reach the goals defined in a pluri-annual programme.

The Laboratory, being part of the Joint Research Centre (JRC), proposes its programme to the General Director of the JRC who - after consultation with a General Consultative Committee - submits it to the Commission which in turn (possibly after adjustments) transmits it to the Council of Ministers.

The Council normally asks for a review by the Group of Atomic Affairs (under the authority of the Permanent Representatives of the member states- the states' ambassadors to the Community in Brussels). The Council then defines the contents of the programme and the requirements in budget and manpower for its execution.

In carrying out the programme thus defined, the Director of the Laboratory is advised by a Consultative Programme Committee, established by the Council of Ministers and including delegates from all member states. It meets three times a year to judge the progress made and to compare it with the original plan. It may, upon request, report to the General Consultative Committee, thus providing information to the Commission and the Council.



Senior staff – contemplating

On the Laboratory scale, the concept of projects has largely replaced the traditional structure, which grouped laboratories of similar nature in divisions such as Physics, Technology and Chemistry. Each project is centered on a major problem which is tackled over a period of years by a team of specialists drawn from a variety of backgrounds.

At present (1975), the following major projects are under way:

1. Swelling of fuels for advanced breeder reactors
2. Corrosion of steel cladding / mixed oxide fuel interfaces
3. Contribution of 5f electrons to bonding in actinides and their compounds.

The following mini-projects are also being studied:

4.1. Ultrasonic thermometry in pile

4.2. Equation of state of plutonium bearing oxides and other nuclear materials

4.3. Burn up and fission yields in fast fluxes.

Some of these projects will probably become a large part of the next pluri-annual programme, scheduled to start on January 1, 1977.

The laboratory practices very detailed forecasting of the development of the work, which is done by means of project flow sheets. These are reviewed annually and revised biannually, and allow a close check on the rate of success.

The responsibility for the execution of each project rests with a speaker (and his deputy), who are proposed by the members of the group. The aim is to achieve defined results within defined time limits, and for this purpose support in the form of personnel and finance is allotted to each group. Within each project group, the individual scientific or technical collaborator reports directly to the project speaker, and is personally responsible for the execution of his particular elements of the total programme.

The various special laboratories (about a dozen) are likewise solidly anchored in the project framework. They each receive budgetary support from the management in proportion to their respective contributions.

The Programme Committee – cheerful

We believe this structure gives the optimum efficiency for a laboratory of the size of the Transuranium Institute. It allows the Institute's programme to be executed in a coordinated and open way, while maintaining an acceptable degree of individual freedom and personal responsibility for each employee.
Moreover, it facilitates the reporting operation and enables the members of the Consultative Programme Committee to verify with relative ease how the allotted funds are used.
A multinational laboratory can be justified on a financial basis if its sponsors benefit from its work **in relation** to their share of the financial burden.
This implies also the execution of medium research and of highly specialized operations which the member states are not willing or able to carry out in their own laboratories.
By periodic technical meetings with the respective departments of the national laboratories and thus by a perpetual exchange of ideas the complementarity and the usefulness of the Laboratory's work is assured.

Some general remarks:
The achievements of a scientific laboratory are difficult to quantify as financial benefits. This is more difficult, the further they are away from any technical realisation.
The MASURCA fuel element production was an essential element in the early development of a fast breeder family which today is considered the leader in this field.
The switch to medium and long term research makes benefit estimations more difficult. Still, a simple argument is convincing:
Large scale energy supply is the major and most costly problem of the industrialised world, measurable in orders of 10^{12} DM/year in direct costs and in specified penalties (in the case of non-supply) of the same order. Nuclear energy, and finally fast breeder energy, will have to take an increasing share.
Fast breeder fuel amelioration, consequently, has financial consequences of considerable magnitude, and the expenses necessary to reach that aim seem to be fully justified.

Administration – moving vigorously

Safety First

The transuranium elements, especially plutonium, are often referred to as the most toxic substances known to man.

They decay by emitting energetic alpha particles which deposit their energy within a very short distance of their point of origin. Thus, although unable to penetrate a sheet of paper, they inflict damage on all living tissues. Damaging effects always occur when alpha-emitting nuclides are incorporated into the human body. In fact, it has been found that the greatest risk originates from inhalation. Protection of people working with transuranium nuclides must be based on adequate control and containment of these materials. In addition, although alpha emitting nuclides generally can be handled at short distances, work with irradiated fuel and with certain transplutonium elements requires heavy shielding against gamma-rays and/or neutrons. The following table illustrates the handling problems of ^{144}Ce, a gamma-emitting fission product, of ^{239}Pu and of ^{252}Cf, an alpha, neutron and gamma emitting transuranium nuclide.

Nuclide	type of radiation	halflife years	shielding required for 1 mg nuclide at 2 m *	maximum permissible conc.in air, mg/m^3*
Ce-144 Pr-144	Gamma	0.78	15 cm concrete	2.10^{-9}
Pu-239	Alpha	24 400	no shielding required	3.10^{-8}
Cf-252	Alpha Gamma Neutrons	2.65	90 cm water	1.10^{-11}

* for a 40 hour week occupational exposure

Safety requirements for some radioactive nuclides handled at the TU Institute

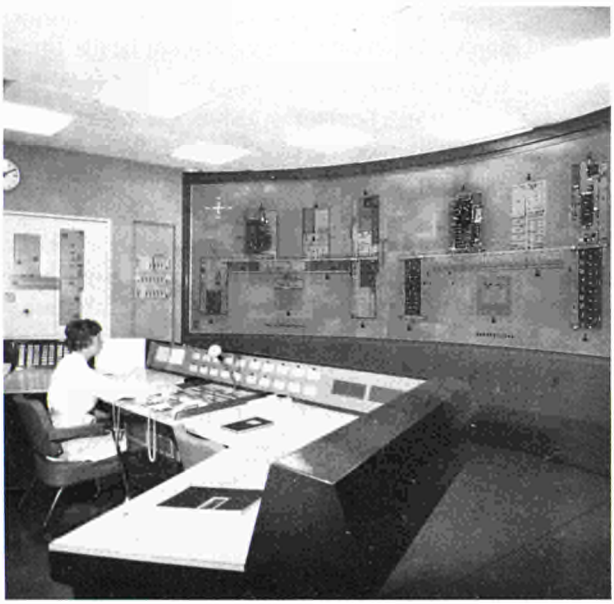
The efficient protection of personnel and of the general public against release of and radiation from transuranium nuclides handled in the Institute are assured by:

- suitable **installations and equipment**
- appropriate **working rules** and **handling techniques**
- the radiological **protection service**.

Installations and equipment

Since control of air purity is of the utmost importance, the buildings of the Institute have been constructed with a view to complete regulation of all air flows. Exchange of air between inside and outside occurs at only a few places and in predetermined quantities. A system of stepwise increasing underpressures forces air entering the buildings to flow in the direction of greater contamination risk. Before leaving the building by way of the exhaust stacks, the air passes through a series of absolute filters. Continuous monitoring of the air leaving these stacks has shown that during the past 5 years the amount of

^{239}Pu given to the atmosphere did not reach 0,3 % of the amount permitted by the authorities. In the hot laboratories, the zone of lower pressure, the unirradiated plutonium is handled in glove boxes which are at a constant underpressure with respect to the room. All equipment used in these glove boxes has been carefully selected and checked for all kinds of safety risks. Work with irradiated plutonium or certain transplutonium elements is carried out in hot cells with telemanipulators. A system of automatic and manual alarms connects the various working places with the control room , which is manned round the clock.



Control room

Working rules and handling techniques

The benefits of good equipment depend fully upon the persons who work with it, and safe handling of transuranium elements is feasible only when workers follow clearly defined rules. Every worker in the laboratory is well aware of the safety aspects of the work he is doing and consequently willing to accept certain inconveniencies caused by the rules and techniques required for working with radioactive substances. Most of these rules are aimed exclusively at the prevention of spills of radioactive material which is handled in each laboratory. In addition, by frequent checking of hands, feet, clothing and equipment for contamination, each worker contributes to the early detection of uncontained radioactivity.

Radiological Protection Service

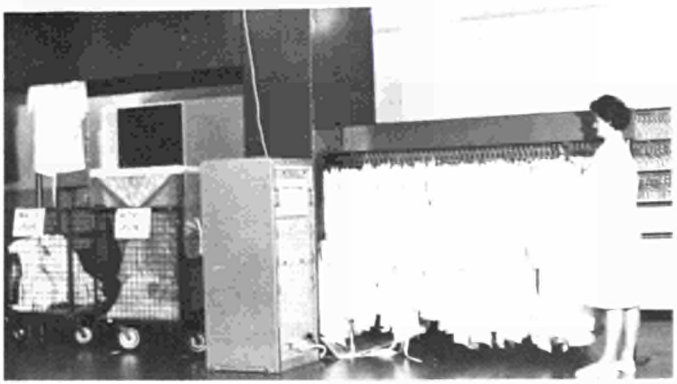
The radiological protection service of the Laboratory, which is fully independent from other technical and scientific groups

- assists in the planning of projects or experiments by advising on safety matters
- supervises the execution of agreed safety measures and following up of working rules
- provides and maintains safety equipment (e.g. gas masks) for the personnel, and instrumentation for checking for contamination and radiation
- carries out a great variety of measurements aimed at checking the contamination of air, laboratories or equipment, and the radiation doses received by each member of the personnel
- is able to organise and supply relief in emergency situations (spills, fires, criticality accidents[1]).

Air monitoring is carried out to watch both for any sudden release of substantial amounts of radioactive materials and for a gradual escape which could give rise to a continuous low level of contamination. Continuous air monitors rapidly detect quantities of radioactivity which exceed the maximum permissible hourly intake. Much more sensitive, but with a delay of eight days, a discontinuous measurement for ^{239}Pu has a limit of detection of 6×10^{-11} mg/m^3. The air monitoring programme is complemented by routine contamination checks of laboratory clothing,shoes, laboratory floors, furniture, equipment and gloves.

The system of alarms mentioned before permits radiological protection personnel to be on the spot within minutes of an incident, so that immediate help is assured.

A final check on the effectiveness of all safety precautions and on each individual worker is provided by regular analyses for alpha emitters in urine and by the continuous measurement of the external radiation dose received by each person.



Laboratory clothes are continuously monitored for contamination

[1] Criticality: A state of affairs in which a sufficient quantity of fissionable material is assembled in the proper shape and concentration for a self-sustaining chain reaction to take place.

Contamination outside hot cells and glove boxes very rarely exceeds natural background, and the average total body exposure of the personnel is less than 3 % of the limit prescribed yearly for radiation workers. These results show that the principle that doses be kept as low as is readily achievable, put forward by the International Commission on Radiological Protection, is put into practice in the Institute for Transuranium Elements.

The following figures demonstrate the activity of the Institute's radiological protection service:

Hand and foot monitors in continuous operation	*40*
Alarm sensors installed in glove boxes and connected to the control room	*1 800*
Number of signals registered per year in the control room	*5 000*
Smear tests executed in laboratories per year	*11 000*
Pieces of clothing handled per year by contamination monitor	*16 000*
Measurements of air sampling filters per year	*45 000*
Floor area checked for contamination per year in m^2	*80 000*

Beyond the Day's Work...

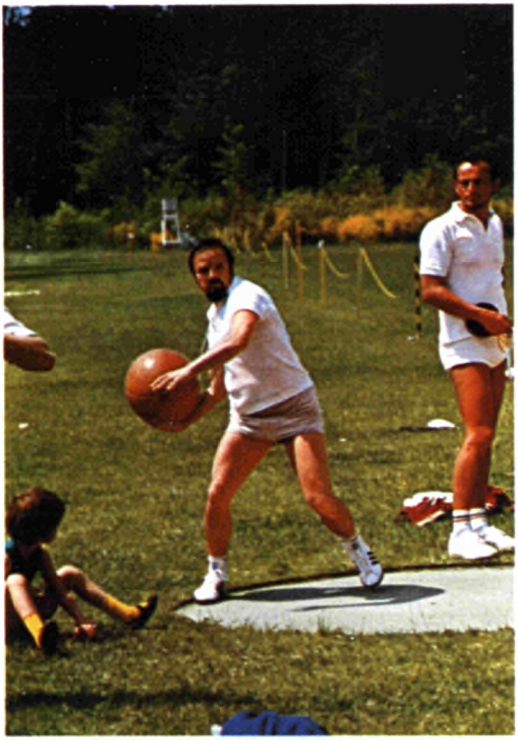
The staff members and the visiting scientists have open to them a large number of social activities. These are, at least partially, financed by the Commission and are managed by a committee (COPAS) which is composed of delegates from the administration and elected representatives of the personnel.

At present COPAS supports twenty different groups whose activities range from judo, fencing, soccer and tennis to chess and stamp collecting. The sports groups take advantage of the excellent facilities of the European School. The football and volleyball teams regularly take part in tournaments with other national and European institutions.

COPAS also arranges seasonal social gatherings such as dances in February, June and November, a general sports day for all members of the Institute and their families, a family excursion on Schuman Day, May 9, and a Christmas party for the children.

Thus there are ample opportunities for the staff members with their families to get together; the children even more so as a result of the summer holiday camps, arranged with financial support from the Community during school holidays.

A variety of possibilities exist for those who wish to broaden their education and improve their abilities. German is taught to newcomers from other countries. Courses in French, English, Italian and Russian are given, and there are lectures on subjects such as Reactor



A sports event

After hours...

Technology, Health Physics, Higher Mathematics, and Solid State Physics ..
All members of the Institute may participate in external courses, evening classes and specialist training programmes which the Commission supports.
The educational programme is rounded off by the Institute's Seminary where interested staff members gather every two weeks to discuss problems of general interest or to listen to invited lectures on topics which range from Fuel Technology, Energy Policy and Ecology to Cryogenics, Astrophysics and the European Implications of Modern Art.

The City with the European Touch

Karlsruhe is situated in the Rhine valley, just below the Black Forest and midway between the Castle of Heidelberg and the Cathedral of Strasbourg. Different stories are told why and how the city of Karlsruhe was founded in 1715 by the Markgraf Karl Wilhelm von Baden-Durlach. The children learn at school that Karl Wilhelm's wife had lost her fan in the forest called Hardtwald. On his attempt to find it, the Markgraf fell asleep under an oak tree and saw in his dream how the fan turned into a fan-shaped city, and he promised to build this city as he had seen it.

The other version has it that the Markgraf's wife was jealous of him, with good reason, which made the Markgraf suffer so much that he left her alone in the Durlach Castle and moved into a newly constructed residence in the Hardtwald, the architecture of which was inspired by Versailles... The city of Karlsruhe, meaning Karl's resting place, then grew around the castle. In his new residence, the Markgraf was surrounded by a female bodyguard, called the Tulipgirls, which sheds some doubt on the justification for the choice of name for the place.

Whatever may have been the reason for founding Karlsruhe, the result impressed Thomas Jefferson so much that he recommended it as a model for the construction of the American capital, Washington, D.C.

Right from the beginning of its history, Karlsruhe was a truly European city:
- It was constructed with the aid of masons and workers from southern Italy, who added a particular note to the local dialect, the Brigande-Deutsch which may still be heard in the streets of old Karlsruhe,
- In 1825 the Karlsruhe Polytechnical School (Technical University) was founded by TULLA according to the model of the Ecole Polytechnique in Paris.
- Since 1955, Karlsruhe has had friendly links with two partner cities, the French city of Nancy and the English city of Nottingham.
- Karlsruhe is the terminal of oil pipelines coming from the Mediterranean and from the Black Sea and hosts several multinational petrochemical firms.

Karlsruhe, finally, is not only the home of the European Institute for Transuranium Elements and of Eurocontrol (an international organisation for the supervision of high altitude aircraft traffic) but also of the first European School in the Federal Republic.

Some dates:
1715 Foundation of the city by Markgraf Karl Wilhelm

1806 Karlsruhe becomes the capital of Grossherzogtum Baden
1825 Foundation of the Technical University of Karlsruhe
1902 Karlsruhe has a population of 100.000
1974 The population of Karlsruhe exceeds 250.000.

The European School of Karlsruhe

Following the initiative of a parents association, the first European School for the children of the employees of the European Communities was founded in 1957 in Luxembourg. Other schools followed in Belgium, in Italy and The Netherlands. After the decision to build the European Institute for Transuranium Elements in Karlsruhe, the European School of Karlsruhe opened its doors in 1962.

The European School system is an attempt to co-educate children of different mother languages. Lessons are taught in four languages (German, French, Italian and Dutch)

The European School at Karlsruhe

according to a common plan. English has been added to the programme recently. To promote mutual understanding some subjects are taught in elementary school classes with children from different countries in a language which is not their mother tongue (the so-called European hours). In the upper high-school level history and geography are given for French and Italian children in German, and for children with German or Dutch as mother language in French. The directors and teachers of the school stem from all countries of the European Communities.

The school comprises a Kindergarten (minimum age for admittance 4 years), an elementary school with five grades (minimum age for admittance 5 years), and a seven grade highschool. The final examination (baccalaureat européen, europäisches Abitur) is recognised as a prerequisite for access to all universities of the member states of the European Communities.

After a very modest start as a guest in the Werner-von-Siemens-Schule, the Karlsruhe European School moved into its new buildings in Karlsruhe-Waldstadt in 1968.

As to be expected, relations between the European School of Karlsruhe and the Institute are very close. The Director of the Institute is a member of the Administrative Council of the school and the Parents Association is run primarily by members of the Institute.

In 1974 there were
310 pupils in the German division, 217 pupils in the French division
225 pupils in the Italian division, 73 pupils in the Dutch division,

with altogether
76 children in Kindergarden, 403 children in elementary school and
346 children in high-school.

Some Results and Achievements

The results of the scientific and technological work carried out at the Institute are described in Semestrial Reports (TUSR, limited distribution), discussed at meetings and conferences, published in Euratom reports and in scientific and technical journals. A condensed bibliography is given at the end of the booklet.

A description of the results obtained at the Laboratory during the ten years of its existence must, in the present context, be incomplete and superficial. Only a selection can be included. More detailed information may be obtained by referring to the bibliography or by contacting the Institute's Documentation Service.

As already indicated, the programmes of the Institute (Plutonium and Transplutonium) are centered on two main subjects:

- **Fuel Science and Technology** (mainly plutonium) and

- **Basic Actinide Research** (mainly transplutonium elements).

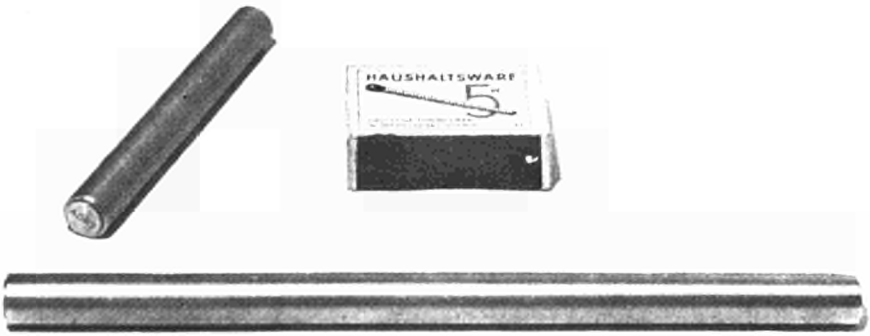
Fuel Science and Technology is to be understood in the broadest sense, comprising the development of fuel fabrication techniques, the study of physical, physicochemical and thermodynamic properties of plutonium-based fuels, the preparation and execution of irradiation experiments, the post-irradiation structural, chemical and isotopic analysis, the study of fuel-cladding interactions and the investigation of swelling phenomena.
The objective of the Basic Actinide Research carried out at the Institute is to understand the chemical bonding in actinide elements and compounds and in particular, the role of 5f electrons in atomic interactions. Work on actinides in general concerns the production, separation and purification of transplutonium elements, the preparation of alloys and compounds, the investigation of their thermodynamic and physical properties and the study of technological applications.
Although this rather crude subdivision has in fact been substituted lately by a more detailed project organisation, a mixed arrangement is used in this review.

Work on these subjects proceeds in various laboratories all over the world, but the European Institute for Transuranium Elements at Karlsruhe offers the unique opportunity of having all these activities under **one** roof and of providing close cooperation between all teams involved.

Fuel Science and Technology

Fuel Preparation and Characterization

In 1967, 2100 metallic fuel rods containing 187 kg of plutonium were fabricated at the Institute for the French zero-power reactor MASURCA. The rods consisted of a Pu-alloy



Fuel rods for MASURCA

with 74 % uranium and 1 % iron. They were prepared by centrifugal casting and clad in stainless steel tubes.

Another subject of major concern is the preparation of mixed oxide powders by mechanical blending and by coprecipitation, and the preparation of sinters with well defined properties and various densities. Sol-gel methods have been developed and used to fabricate spherical particles as starting material for the production of fuel compacts. Powder vibro compaction techniques have been studied and used for the fabrication of several test pins.

For the preparation of carbides and carbonitrides, the influence of compact size of the reaction pellet on the degree and rate of reaction has been investigated in detail.

A fuel pin with annular geometry has been designed which should help to increase the average lifetime of a mixed oxide fuel rod.

Besides the MASURCA fuel, a large number of oxide fuel specimens and some metallic samples have been prepared upon request for laboratories in the community member states.

The chemical composition, especially the impurity content of specimens thus prepared

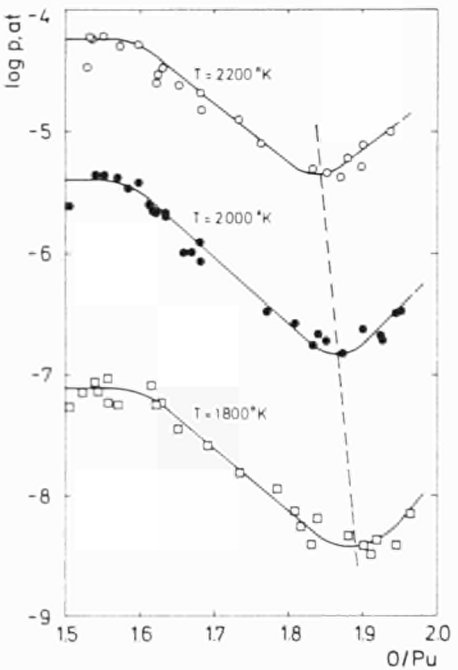
for studies of properties or irradiation experiments are determined by electrochemical, spectrochemical, gas-analytical and gravimetric methods which had to be adapted to the particular properties of the heavy elements and their compounds. Several procedures, such as the titration of plutonium solutions, the determination of carbon and the evaluation of emission spectra have been fully or partly automated.
Some of the analytical methods developed at the Institute are applied within the framework of an international programme on nuclear safeguards.

Studies of Fuel Properties

Valuable information on the appropriateness of a particular fuel for long-term missions in fast power reactors, on its technical feasibility , and on its safety aspects can be obtained by out-of-pile studies on unirradiated samples of fuel material.

Thermodynamics

In this context, thermodynamic properties such as vapour pressure, heat of sublimation and melting points (liquidus-solidus surfaces) of oxide fuels and of advanced fuels are of interest.



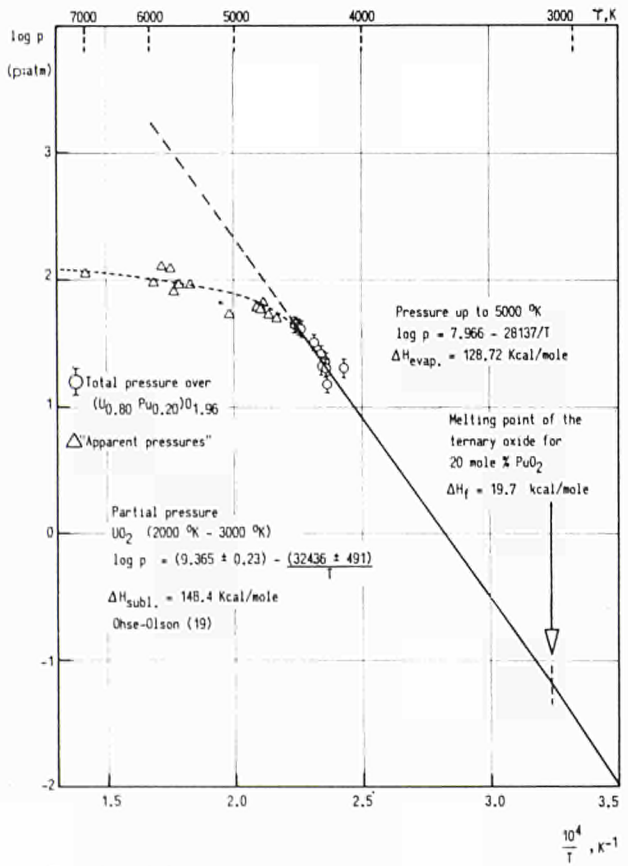
Vapour pressure isotherms at 1800 K, 2000 K and 2200 K versus composition of substoichiometric plutonium oxide

KNUDSEN and torsion effusion methods combined with target collection and mass spectrometric ion intensity measurements have been applied at the Institute to vapour pressure studies.

The vapour pressure of UO_2 has been measured in the temperature range 2200 - 2800 K, giving evidence for oxygen loss of the oxide under vacuum at high temperatures. The data have been used in extrapolation to higher temperatures. The bivariant behaviour of PuO_{2-x} in the single phase region down to Pu/O = 1.61 has been investigated. The congruently evaporating composition was determined as a function of temperature.

The vapour pressures of the mononitrides (U,Pu)N have been studied.

Previously, vapour pressure data for the solid range had been available only up to temperatures of 3000 K for UO_2 and 2300 K for $(U,Pu)O_2$. As extrapolations over several thousand degrees can only be a rough guide, an extension to higher temperatures was urgently required.



$(U_{0.8}Pu_{0.2})O_{1.96}$ vapour pressure data up to 7000 K

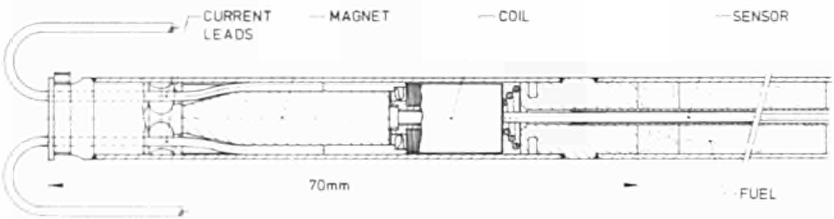
The development of a new laser surface heating technique combined with fast optical pyrometry allowed vapour pressures to be measured up to 5000 K.
A new technique for phase transition analysis by mass spectrometric ion intensity measurement has been developed, which allows microanalysis and the measurement of materials of low thermal conductivity The monocarbide (U,Pu)C system and the ternary $(U,Pu)O_2$ oxide system have been investigated in this way.

Thermal Properties

Knowledge of fuel thermal characteristics is important for the evaluation of fuel behaviour under irradiation. For the interpretation of the structural features of irradiated fuel , information is needed about the temperature distribution across a section of a fuel pin, and the central temperature in a pin at a given heat load is one of the principal parameters for the evaluation of the safety of a particular configuration of a reactor core.
Therefore, continued emphasis is laid on:

- the study of the thermal conductivity of non-irradiated and on burn-up simulated mixed oxides, carbides, nitrides and carbonitrides,
- in-pile measurements of the central fuel temperature in oxide fuel pins
- attempts to measure fuel-to-cladding contact conductances during irradiation.

Stoichiometric $(U,Pu)O_2$ was found to be a better heat conductor than non-stoichiometric mixed oxide. The addition of solid fission products decreased the thermal conductivity of mixed oxides in a pronounced way at temperatures below 1000°C.
A new type of temperature sensing device, based on the measurement of the temperature dependent velocity of sound in an appropriately shaped sensor has been developed and



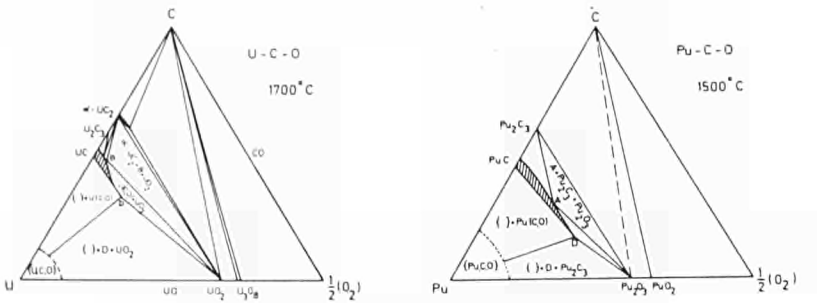
Thermometer capsule for the TRESON-1 irradiation experiment

tested. It will be used for measuring in-pile central fuel pin temperatures above 2200°C and for prolonged irradiation periods.
The thermal conductivity of mixed uranium-plutonium carbides, carbonitrides and nitrides has been investigated systematically. For the temperature region between 800°C and 1500°C empirical relations have been obtained to predict the conductivity of samples with known compositions.

Phase Diagrams

Knowledge of the mutual solubilities of the constituents of multicomponent systems and

of the variation of melting temperatures with composition is a prerequisite for the development of fuel fabrication methods as well as for the understanding of phenomena accompanying constitutional changes in a (multicomponent) fuel material.
This is why the phase diagram of the U-Pu-O system , for example, has been studied in detail. In particular, the U_3O_8-phase and the influence of Pu-addition on its extent has been investigated. Relations between the U_4O_9-phase and U-Pu-O-phases with the same oxygen content were established. The solubility limit of plutonium in U_3O_{8-x} at various temperatures was determined as well as the oxidation limit for mixed oxides at 1 atmosphere of oxygen.
Furthermore, in mixed oxides tetragonal phases with O/M-ratios around 2.3 have been investigated.
The variation of the lattice parameters of mixed oxides with plutonium and oxygen content and of mixed carbonitrides with nitrogen content has been determined. Two phases were found in hypostoichiometric oxides with a Pu-content above 20 %.



The U-C-O and Pu-C-O ternary diagrams

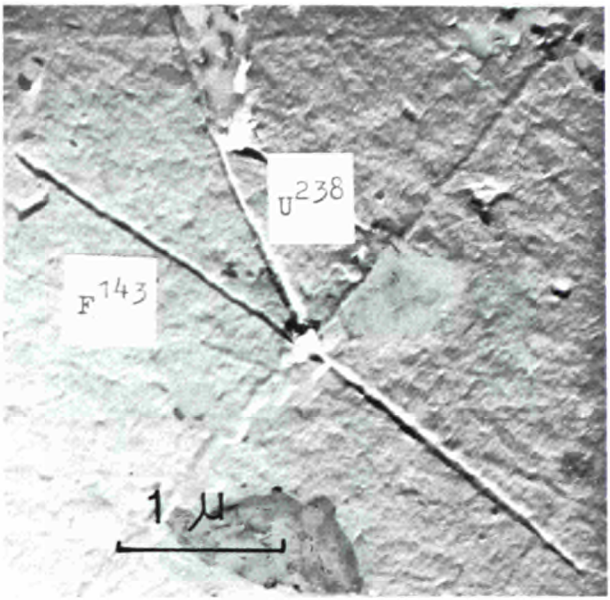
Studies of phase relationships in the U,Pu-C-O and U,Pu-N-systems have been performed in regions of technologically relevant composition.

Study of Lattice Defects

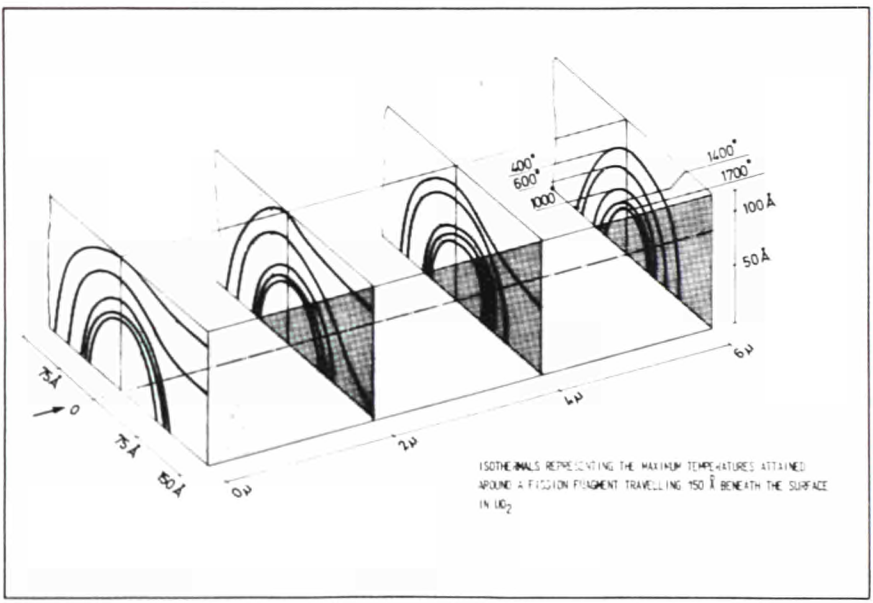
Transmission and replica electron microscopy have been extensively applied to the study of structural defects such as dislocations and to fission spikes and their effects on macroscopic phenomena in irradiated and unirradiated oxide and carbide fuels.

The properties of dislocations in unirradiated UO_2 and UO_{2+x} and in particular the differences in glide properties of dislocations belonging to different crystal planes have been investigated in connection with plastic deformation experiments at high temperatures.

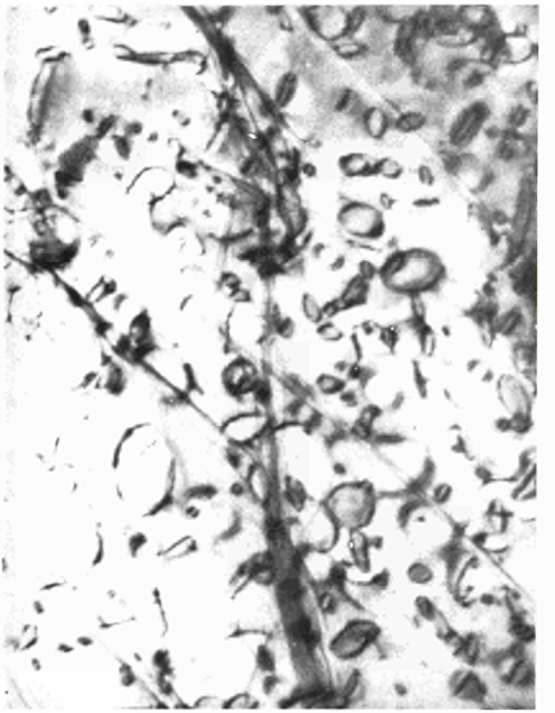
The effect of neutron irradiation on fuel plasticity has been studied in connection with the in-pile creep experiment SOCRATEM. It was found that under irradiation the mobility of glide dislocations was reduced due to the formation of interconnections and jogs, which lead to a hardening of the material.



Scattering event between a fragment of mass 143 and an oxygen lattice atom



Isotherms representing the maximum temperatures around a fission fragment travelling 150 Å beneath the surface of a UO_2 sample



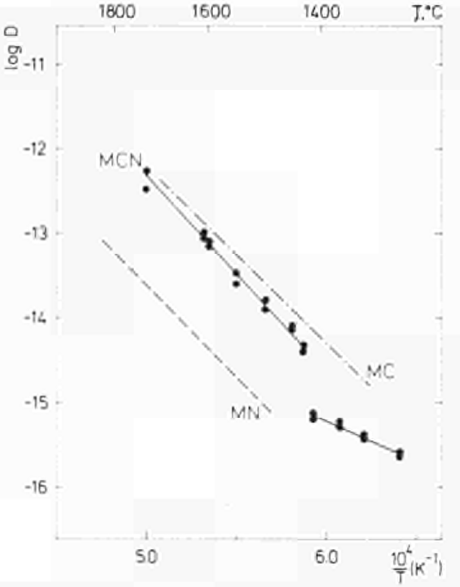
SOCRATEM I. Electron-microscopic view of the glide plane of the creep sample (Large antiparallel components of the Burgers vectors have formed a dipole at a)

Actinides and their compounds are submitted to continuous self-irradiation which, in the case of solids, creates lattice defects and thus affects certain physical properties of these materials. The influence of self-irradiation on the interatomic distances in PuO_2, AmO_2, $CmBe_{13}$ has been studied extensively. These studies are of particular importance for the correction of lattice parameter determinations on unannealed samples.

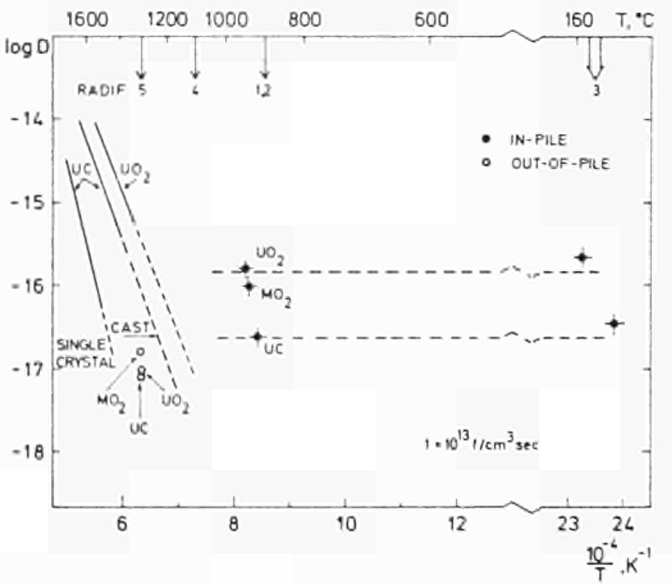
Diffusion studies

In the oxides, carbides and nitrides of uranium and plutonium, the metal atoms are much less mobile than the oxygen, carbon or nitrogen atoms. The diffusion of U and Pu can therefore be rate- controlling for many high temperature kinetic processes, such as grain growth, sintering, creep, void formation and swelling.

In addition, diffusion coefficients are needed as input data for nuclear fuel performance codes. Therefore, diffusion studies with U and Pu were performed to provide the missing data and to explain discrepancies existing in the literature.



Arrhenius diagram for the diffusion of ^{238}Pu in (U,Pu)C, (U,Pu)CN and (U,Pu)N



Arrhenius diagram for radiation enhanced diffusion in UO_2, UC and (U,Pu)O_2

High resolution α-spectroscopy was used to measure diffusion coefficients, and this method has been developed both experimentally and mathematically to such a perfection that the effect of many possible parameters (small or large deviations from stoichiometry, small or large impurity additions, different grain size) could be investigated accurately thus explaining most of the scatter in literature results. Separate experiments performed in a nuclear reactor at temperatures between ambient temperature and 1300 °C, besides providing the temperature functions for thermally activated diffusion between about 1000 °C and the melting point also yielded the effect of radiation environment and fission on atomic mobilities, hence the radiation enhanced diffusion.

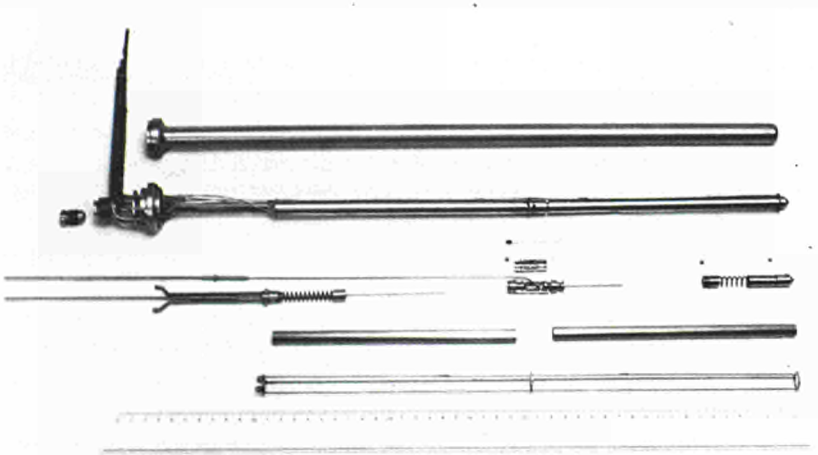
This work has resulted in sets of diffusion coefficients for both U and Pu in MO_2, MO_{2+x}, MO_{2-x}, MC, MCN, MCON and MN with M being either (U + Pu) or U alone. The results served as a basis to develop models of the atomic defect structure and they could frequently be correlated with the above-mentioned kinetic processes.

Study of Fuel under Irradiation

Design and Execution of Irradiation Experiments

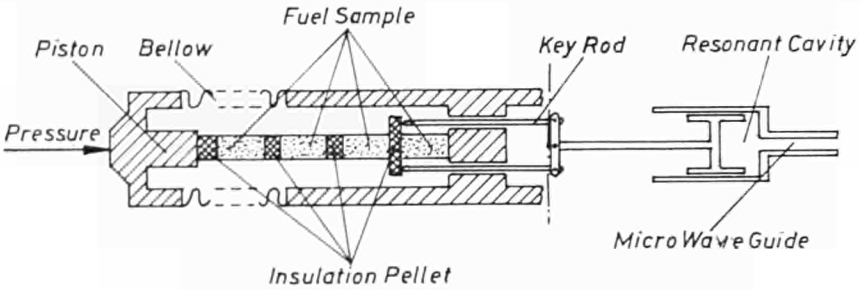
The study of the behaviour of fuels and fuel pins under irradiation is a necessity for basic fuel research and is often of immediate technological relevance.

Numerous irradiation experiments have been prepared, executed and analysed at the Laboratory. Some of them were concerned with the influence of density, stoichiometry and fabrication procedure on the in-pile behaviour of mixed oxide fuel pins in a fast neutron flux (the DS and DD-experiments). Others were aimed at comparing mixed carbides,



Assembling the MIN-UP II irradiation device

nitrides and carbonitrides in a fast flux at various irradiation levels, especially with regard to their swelling properties (the DN-experiments).
A unique instrument, the POM-capsule, was developed in order to irradiate fuel samples of various types in a relatively short time to a high burn-up (values of 13 a/o were attained after 80 days of irradiation in the BR-2 reactor at Mol, Belgium).

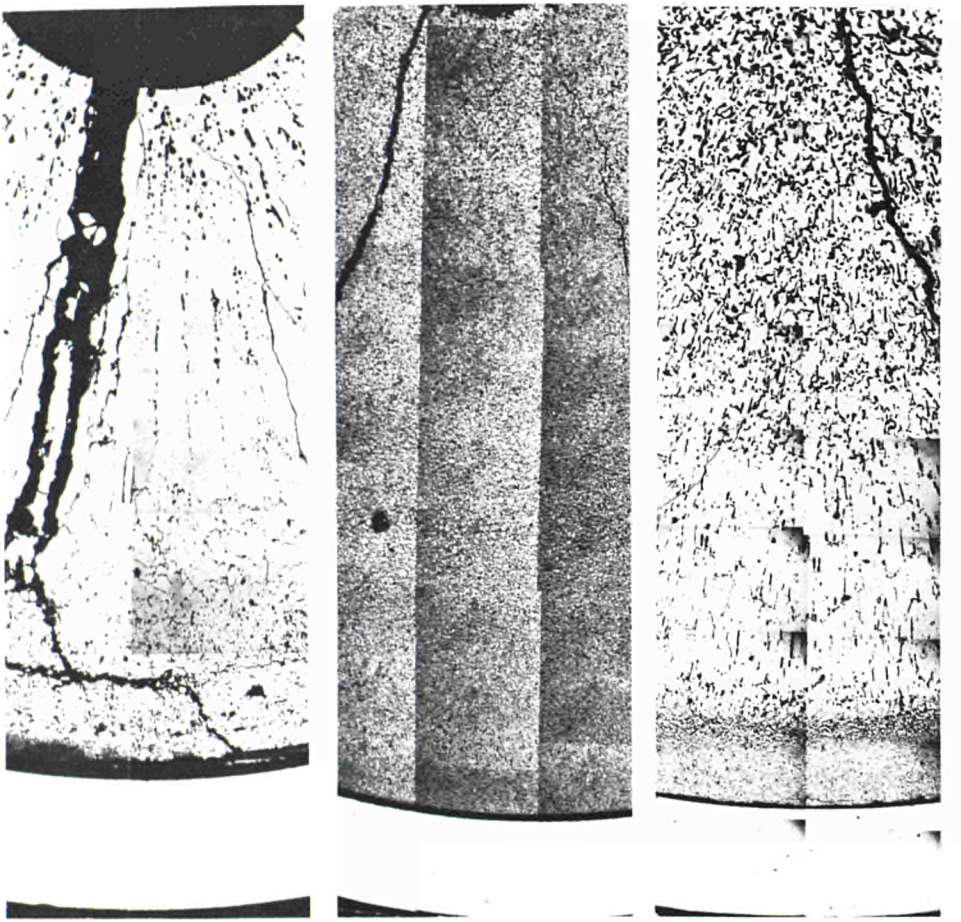


Schematic view of the SOCRATEM irradiation device

Several instrumented devices have been designed or adapted and irradiated in different reactors to investigate particular phenomena such as the evolution of central temperatures in mixed nitride fuel (MINUP), the influence of irradiation on the mechanical deformation of fuel under load (SOCRATEM), on diffusion processes (RADIF), on the swelling of carbides (GOCAR) and the applicability of ultrasonic methods to the measurement of centre line temperatures of oxide fuel pins (TRESON).

Post-irradiation Structural Analysis

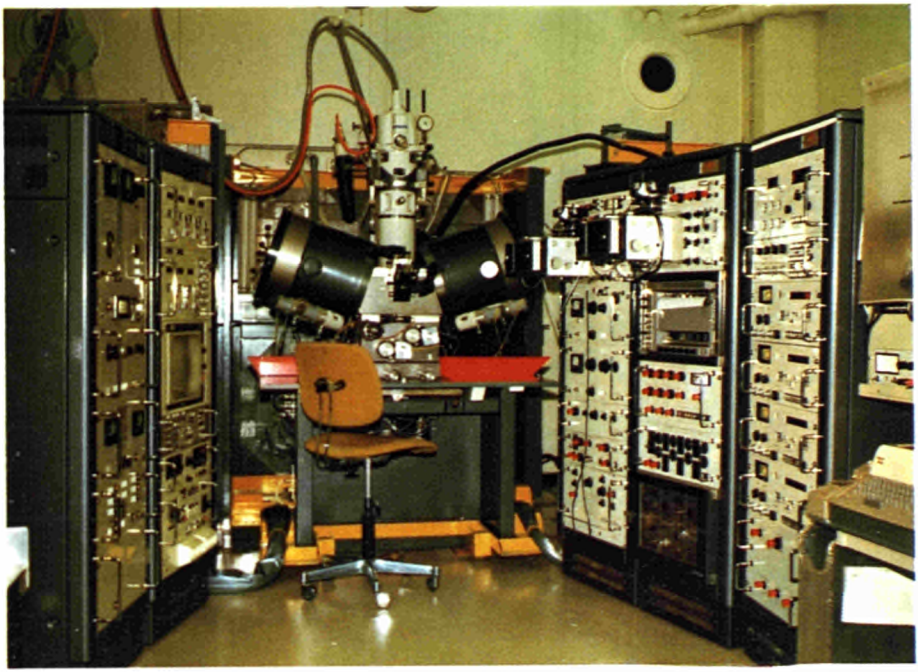
After irradiation, fuel pins or capsules are returned to the Institute and examined in alpha-gamma-tight hot cells or with appropriately shielded instruments.
Dimensional measurements, radiography, fission gas analysis, alpha- and beta-gamma-autoradiography, microscopic investigation of fuel structures, lattice parameter measurements on micro-samples, electron-microscopic examination of fuel replicas, and the determination of local chemical composition by electron microprobe analysis are the principal techniques employed at the Laboratory for the examination of irradiated fuel specimens.
They permit the investigation of the important consequences of high central fuel temperatures and steep thermal gradients on the fuel structure and on the migration of fuel constituants and fission products in oxide fuels. This migration may lead to an attack on the cladding material.
It was found that 90 to 95 % of the fission gas is released from the oxide fuel during irradiation at high heat ratings and that the swelling of oxide is almost entirely due to the presence of solid fission products. It amounts to about 0,9 % per a/o burn-up.



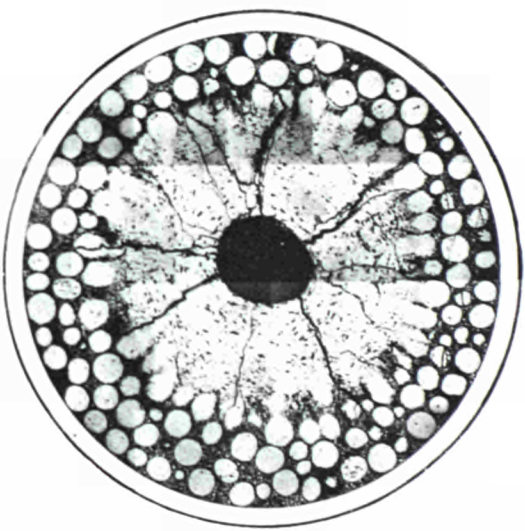
(a) (b) (c)

Microstructures of fast flux test pins – a) Mixed oxide fuel (DS2), O/M = 1.97, burn-up 7%; b) Mixed nitride fuel (AP3), burn-up 1.3%; c) Mixed carbide fuel (DP2), burn-up 1.3%

This, of course, is different in the case of advanced fuels, where the amount of fission gases released remains below 65 %. When these materials are exposed to high heat loads (1350 W/cm, 2200°C central fuel temperature), coaxial grains start to grow, columnar grains are formed, interconnected pores appear and sintering occurs. These phenomena depend on the chemical composition of the fuel and on the mobility of the fuel constituents.



The shielded microprobe analyser CAMECA Ms 46



GIANO 2, restructuring of sol-gel fuel after 12 hours of irradiation

Restructuring and geometrical swelling were found to be less accentuated in mixed nitrides than they are in carbonitrides and in carbides. The averaged geometrical swelling rate of nitrides was 0.12 % per a/o burn-up.

The migration of plutonium, uranium and of fission products in advanced fuels under irradiation does not seem to cause corrosion problems comparable to those observed with oxides, whereas carbon may promote corrosion in the case of carbides.

In eight years of operation, the Hot Laboratory examined 141 irradiated fuel pins and ten irradiated capsules of which 80 pins and all capsules underwent complete destructive examination. The capsules as well as 20 oxide pins and 15 pins containing advanced fuels had been irradiated within the framework of the Institute's own research program. The other examinations were carried out upon request from laboratories in the member states of the European Communities.

Simulation Experiments

The post-irradiation work is complemented by simulation experiments, where non-irradiated fuel samples are submitted to conditions which might prevail during irradiation by adding non-active fission product elements to the fuel matrix and/or applying temperature fields which are characteristic for fuel pins in operation. A better knowledge of the redistribution of oxygen, of plutonium and of certain fission products as well as of the formation and development of structural features during irradiation has been obtained by these experiments.

Uranium-plutonium carbides, nitrides and carbonitrides with fission product additives in amounts simulating burn-ups between 3 a/o and 17 a/o were prepared and investigated by ceramography, by x-ray diffraction and by means of the electron microprobe. The swelling rate due to the accumulation of solid fission products in mixed carbide fuels was deduced from these investigations to be 0.49 % per a/o. The diffusion of plutonium in these substances was studied, and their heat conductivities were measured at temperatures between 800°C and 1500°C.

Swelling of Advanced Fuels

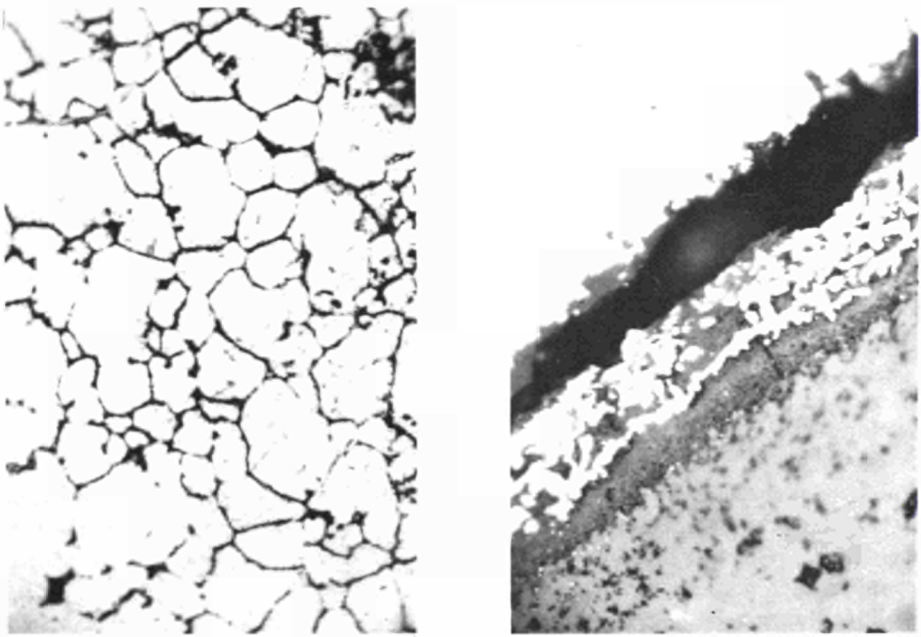
Within the framework of the Laboratory's program on swelling, rods of mixed carbides, oxycarbides, carbonitrides, and nitrides were irradiated at a maximum linear power of about 1350 W/cm at the Dounreay and Rapsodie fast reactors and analysed with special regard to alterations of the fuel structure during burn-up.

In all fuels, fission gas bubbles were studied by replica electron microscopy. The results of a detailed structural analysis combined with those of laboratory experiments on self-diffusion, grain growth, pore mobility, thermal conductivity and burn-up simulation have been used to gain information on the mechanisms of fission gas swelling and gas release. Highly rated carbides and carbon-rich carbonitrides exhibited a gas release of up to 65

% after 1 a/o burn-up, whereas the gas release was clearly lower from nitrides and nitrogen-rich carbonitrides.
At medium burn-up (3 to 5 a/o) the gas release rate was somewhat lower for MC and $MC_{0.5}N_{0.5}$ than at 1 a/o burn-up. Larger fractions of fission gas were released from He-bonded pins during the initial (free-swelling) period and up to medium burn-ups than in the case of Na-bonded pins.
The rate of growth of fission gas bubbles under LMFBR conditions was calculated, taking into account fission gas production and precipitation, fission fragment resolution, gas release and the production and precipitation of vacancies and interstitials.
It was concluded that a lower rate of swelling could be achieved in He-bonded carbide-type pins by following a suitable start-up procedure and running them at an optimum range of working temperatures.

Corrosion Studies

Another serious problem connected with the use of uranium-plutonium oxides as a nuclear fuel in fast power reactors is the corrosion of the inner cladding surface during irradiation. This may lead to a dangerous reduction of the thickness of the cladding wall and eventually to a rupture of the cladding with subsequent contamination of the coolant by plutonium and highly active fission products.



(a) (b)

SALAMI II, intergranular clad corrosion (a) and mixed ceramic and metallic phases on the fuel surface (b)

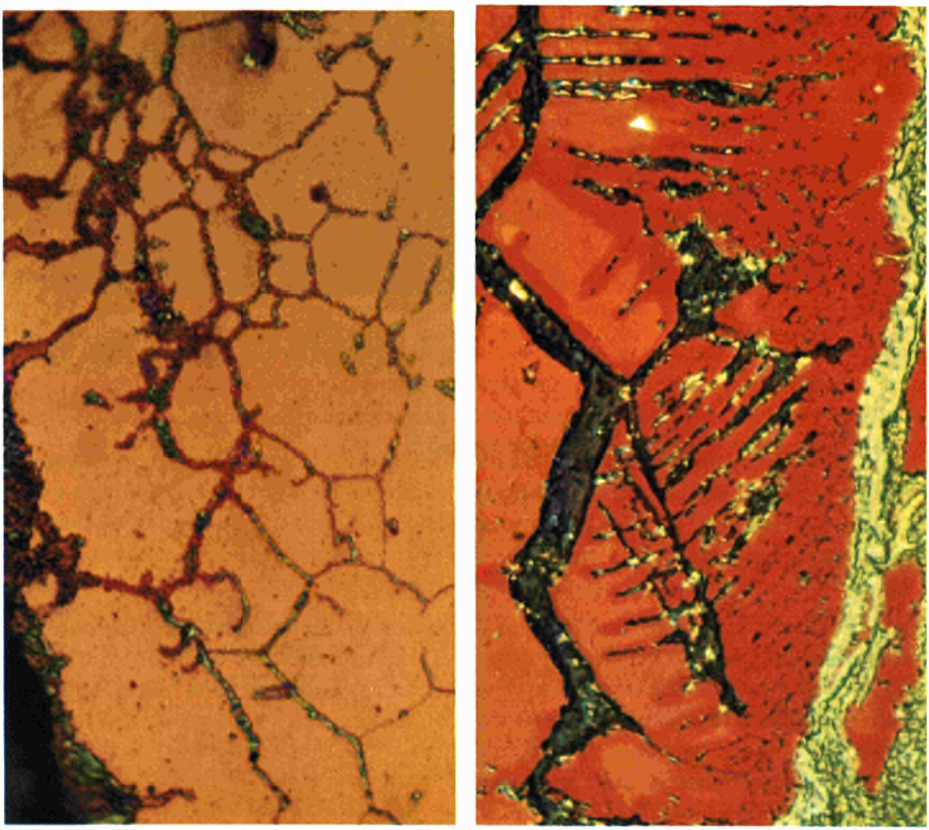
The Laboratory has devoted a considerable part of its activities to the investigation of the processes leading to clad corrosion under irradiation conditions with the hope of finding appropriate means of keeping this corrosion process under control.

The phenomena leading to cladding corrosion under irradiation are rather complex. They consist of

- the transport of the agressive species to the fuel-clad interface under the action of thermal, chemical and electrical driving forces
- reactions of the agressive species with the steel components.

The studies carried out at the Institute are aimed at

- identifying the various steps of the corrosion process and understanding their respective importances
- providing an overall representation of the corrosion process by means of an appropriate model which describes quantitatively the observed phenomena and predicts their evolution with time.



Micrographs of samples of corroded steel cladding which illustrate the use of interference films to enhance phase differentiation

Post-irradiation examination of pins irradiated in the Dounreay and Rapsodie fast reactors has shown that cladding corrosion depends mainly on initial fuel stoichiometry, cladding temperature and burn-up. Corrosion was found to increase as the initial oxygen to metal ratio of the fuel was raised in the range O/M = 1,93 to 2,00. The depth of corrosion was found to increase with increasing burn-up provided the cladding temperature was above 500°C. Increasing Pu-content of the fuel from 20 to 30 % had a negligible effect on corrosion. The different types of cladding material used, 1.4988 and 1.4970 stainless steels and 625 inconel nickel based alloy, did not differ greatly in their corrosion behaviour.

The inner surface of stainless steel cladding has been found to become hardened and embrittled during irradiation, and the amounts of corrosion products in the fuel/cladding gap after irradiation did not correspond to the amount of corrosion that had occurred.

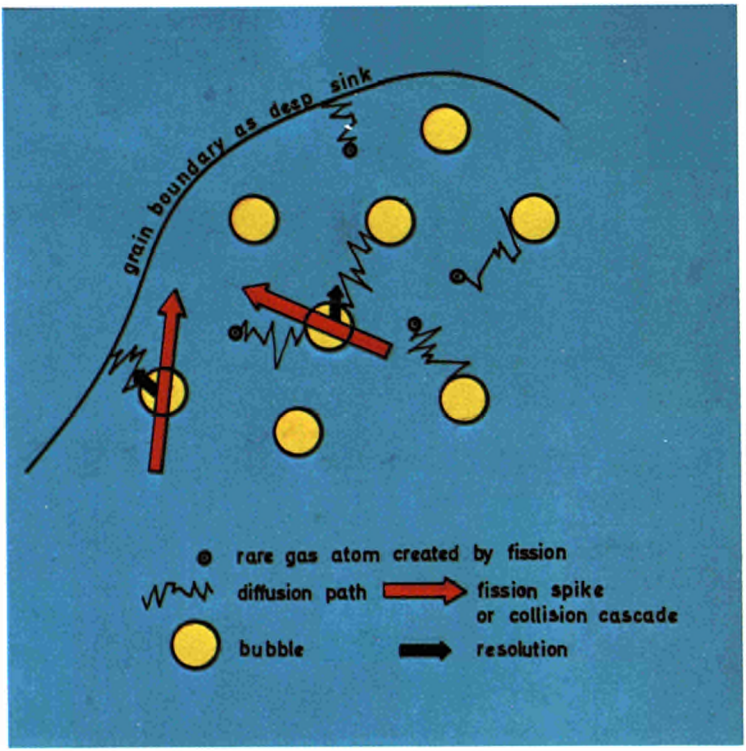
Methods are being developed and applied to measure the radial distribution of oxygen and of certain fission products,such as caesium and tellurium, in oxide pellets that have undergone irradiation. For this purpose, microsamples were drilled out of irradiated pellets in a hot cell and analysed afterwards.

In an attempt to understand and to interpret the various processes leading to clad corrosion, consideration is given to oxide and spinel formation with special attention being paid to chromium depletion and to the properties of passivating layers, the various methods of corrosion attack or ablation, formation of layered structures and most important of all, grain boundary attack. The oxygen potentials and constitutional diagrams of the various oxides of the steel constituents and fission products have been evaluated. The problem of the appearance of gas transport reactions of steel components in the presence of caesium excess was experimentally solved and led to the suggestion of an electro-chemical model for the reaction mechanism.

Basic out-of-pile studies are at present being performed to ellucidate in detail the complex reaction mechanisms under the simulated reactor operational conditions. Electrochemical methods, such as potentiostatic or galvanostatic techniques, as well as electron probe microanalysis and x-ray diffract ometry, are used.

Modelling Work

Parallel to the experimental and theoretical research on fuel materials, the Institute pursues a modelling activity which brings together the various pieces of information on the thermal and structural behaviour of fuel pins under irradiation as parts of a complex system. The manifold interactions of the various parameters of this system and their feedback chains are analysed by means of computer codes. The aim of this work is to evaluate the relative importance of the various components of the system in producing certain phenomena (such as swelling, central melting, cladding rupture, clad corrosion) and to predict the evolution of temperature and of structural features of a given fuel pin configuration under well defined conditions as a function of irradiation time.



Model for the study of bubble migration and resolution in the presence of fissioning atoms

Isotope Analysis

Isotope analysis techniques have been used to determine the isotopic composition of the heavy nuclides and certain fission products produced during irradiation of a given fuel rod and of the burn-up attained. For this purpose, new analytical instruments have been developed which are largely automatic or semi-automatic, e.g., an installation for the separation of uranium and plutonium and mass- and alpha-spectrometers. The resulting data are reduced and analysed by means of computers.

Another application of isotope analysis is the determination of integral cross sections and fission yields in fast reactors (project TACO). These data are of importance for the estimation of the expected lifetime of a fuel rod and of the amount and the composition of the radioactive waste produced in a reactor in a given time.

Relations between isotope-ratios and certain fuel parameters have been determined and the results of this isotope correlation technique applied to fuel management problems and to procedures for supervising the flow of fissile materials through fabrication plants, reactors and reprocessing installations.

ELEMENT	Abundance of Fission Elements (a/o) at Reactor shut-down				
	L W R - 1	L W R - 2	L W R - 3	F B R - 1	F B R - 2
Zn	$7.02\ 10^{-8}$	$1.18\ 10^{-7}$	$1.64\ 10^{-7}$	0.0	0.0
Ga	$2.12\ 10^{-8}$	$3.56\ 10^{-8}$	$4.96\ 10^{-8}$	0.0	0.0
Ge	$1.43\ 10^{-3}$	$1.56\ 10^{-3}$	$1.70\ 10^{-3}$	$1.53\ 10^{-5}$	$3.85\ 10^{-6}$
As	$4.45\ 10^{-4}$	$5.06\ 10^{-4}$	$5.67\ 10^{-4}$	$5.22\ 10^{-5}$	$1.31\ 10^{-5}$
Er	$2.16\ 10^{-1}$	$1.94\ 10^{-1}$	$1.74\ 10^{-1}$	$3.40\ 10^{-1}$	$1.68\ 10^{-1}$
Br	$8.98\ 10^{-2}$	$8.47\ 10^{-2}$	$8.06\ 10^{-2}$	$1.22\ 10^{-1}$	$6.56\ 10^{-2}$
Kr	$1.74\ 10^{0}$	$1.34\ 10^{0}$	$9.64\ 10^{-1}$	$2.09\ 10^{0}$	$1.18\ 10^{0}$
Rb	$9.43\ 10^{-1}$	$6.99\ 10^{-1}$	$4.71\ 10^{-1}$	$1.12\ 10^{0}$	$5.84\ 10^{-1}$
Sr	$3.88\ 10^{0}$	$2.84\ 10^{0}$	$1.88\ 10^{0}$	$3.81\ 10^{0}$	$1.96\ 10^{0}$
Y	$1.76\ 10^{0}$	$1.29\ 10^{0}$	$8.61\ 10^{-1}$	$2.01\ 10^{0}$	$1.08\ 10^{0}$
Zr	$1.25\ 10^{1}$	$1.07\ 10^{1}$	$9.07\ 10^{0}$	$1.34\ 10^{1}$	$9.94\ 10^{0}$
Nb	$1.34\ 10^{-1}$	$1.23\ 10^{-1}$	$1.12\ 10^{-1}$	$1.65\ 10^{-1}$	$1.34\ 10^{-1}$
Mo	$1.08\ 10^{1}$	$1.06\ 10^{1}$	$1.05\ 10^{1}$	$1.12\ 10^{1}$	$1.04\ 10^{1}$
Tc	$2.80\ 10^{0}$	$2.80\ 10^{0}$	$2.80\ 10^{0}$	$2.59\ 10^{0}$	$2.56\ 10^{0}$
Ru	$7.14\ 10^{0}$	$8.56\ 10^{0}$	$9.87\ 10^{0}$	$7.69\ 10^{0}$	$1.07\ 10^{1}$
Rh	$1.53\ 10^{0}$	$1.87\ 10^{0}$	$2.19\ 10^{0}$	$1.82\ 10^{0}$	$2.83\ 10^{0}$
Pd	$4.53\ 10^{0}$	$7.01\ 10^{0}$	$9.29\ 10^{0}$	$2.92\ 10^{0}$	$6.95\ 10^{0}$
Ag	$2.24\ 10^{-1}$	$3.82\ 10^{-1}$	$5.21\ 10^{-1}$	$3.02\ 10^{-1}$	$8.79\ 10^{-1}$
Cd	$1.37\ 10^{-3}$	$2.21\ 10^{-2}$	$2.95\ 10^{-2}$	$1.64\ 10^{-2}$	$3.78\ 10^{-2}$
In	$8.05\ 10^{-1}$	$1.12\ 10^{-1}$	$1.40\ 10^{-1}$	$1.47\ 10^{-1}$	$2.64\ 10^{-1}$
Sn	$1.43\ 10^{-2}$	$1.98\ 10^{-1}$	$2.49\ 10^{-2}$	$3.28\ 10^{-1}$	$2.44\ 10^{-2}$
Sb	$4.24\ 10^{0}$	$6.16\ 10^{-2}$	$7.99\ 10^{0}$	$1.18\ 10^{-1}$	$7.62\ 10^{0}$
Te	$1.27\ 10^{0}$	$1.36\ 10^{0}$	$1.45\ 10^{0}$	$1.51\ 10^{0}$	$1.61\ 10^{0}$
J	$6.80\ 10^{-1}$	$7.71\ 10^{-1}$	$8.58\ 10^{-1}$	$9.11\ 10^{-1}$	$8.94\ 10^{-1}$
Xe	$1.39\ 10^{1}$	$1.41\ 10^{1}$	$1.43\ 10^{1}$	$1.10\ 10^{1}$	$1.15\ 10^{1}$
Cs	$9.07\ 10^{0}$	$9.18\ 10^{0}$	$9.27\ 10^{0}$	$9.22\ 10^{0}$	$1.00\ 10^{1}$
Ba	$3.19\ 10^{0}$	$3.02\ 10^{0}$	$2.86\ 10^{0}$	$3.43\ 10^{0}$	$3.19\ 10^{0}$
La	$2.88\ 10^{0}$	$2.78\ 10^{0}$	$2.68\ 10^{0}$	$2.99\ 10^{0}$	$2.88\ 10^{0}$
Ce	$6.58\ 10^{0}$	$6.22\ 10^{0}$	$5.89\ 10^{0}$	$6.83\ 10^{0}$	$6.39\ 10^{0}$
Pr	$2.47\ 10^{0}$	$2.47\ 10^{0}$	$2.48\ 10^{0}$	$2.72\ 10^{0}$	$2.54\ 10^{0}$
Nd	$8.20\ 10^{-1}$	$7.69\ 10^{-1}$	$7.21\ 10^{-1}$	$8.52\ 10^{-1}$	$7.46\ 10^{-1}$
Pm	$5.93\ 10^{0}$	$5.83\ 10^{0}$	$5.73\ 10^{0}$	$6.95\ 10^{0}$	$7.45\ 10^{0}$
Sm	$1.99\ 10^{-1}$	$2.15\ 10^{0}$	$2.30\ 10^{-1}$	$1.49\ 10^{-1}$	$1.98\ 10^{0}$
Eu	$1.55\ 10^{-1}$	$1.99\ 10^{-1}$	$2.39\ 10^{-1}$	$1.59\ 10^{-1}$	$2.60\ 10^{-1}$
Gd	$1.64\ 10^{-3}$	$2.40\ 10^{-3}$	$3.10\ 10^{-2}$	$7.79\ 10^{-3}$	$1.45\ 10^{-3}$
Tb	$4.96\ 10^{-3}$	$8.15\ 10^{-3}$	$1.09\ 10^{-2}$	$3.42\ 10^{-3}$	$9.36\ 10^{-3}$

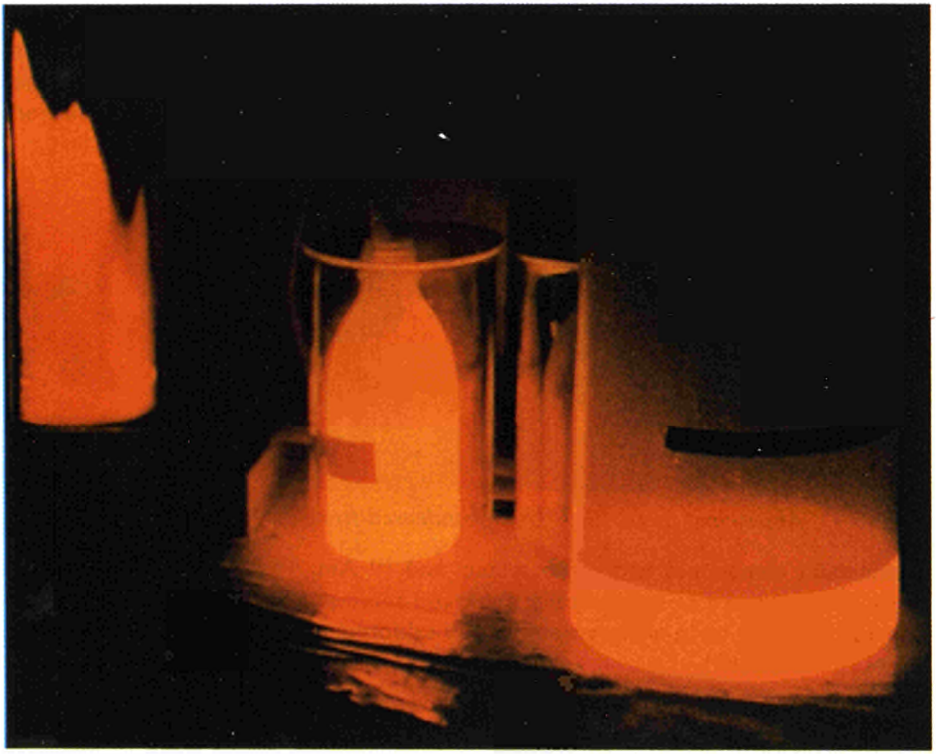
Fission element abundances in different fuels

Basic Actinide Research

Isolation and Purification of Transplutonium Nuclides

Transplutonium nuclides have been isolated from several sets of irradiation targets after exposure of $^{241}AmO_2$-Al cermets to integrated neutron fluxes of 10^{21} - 10^{23} n/cm^2. The transplutonium elements were separated from cladding and matrix material, fission products and plutonium by precipitation, ion exchange and extraction chromatography. For the first time in Europe, microgram amounts of ^{252}Cf were isolated and encapsulated as a spontaneous fission neutron source. The mixture of ^{241} Am and ^{242} Cm oxides was recovered from irradiated targets to fuel a radionuclide battery. After the radioactive decay of isotopically pure ^{242} Cm, biomedical grade ^{238} Pu could be separated.

Multigram amounts of commercial grade ^{241} Am, ^{243} Am and ^{244} Cm oxides have been purified as starting material for the preparation of metals and compounds. Column extraction chromatography with quaternary ammonium salts and acidic phosphorous esters was the most effective method.



Radioluminescence of ^{244}Cm. Separation of multigram amounts of ^{243}Am and ^{244}Cm by extraction chromatography.

Preparation of Actinide Metals and Compounds

Actinium, americium, curium and californium metals have been prepared by reduction of their oxides with lanthanum or thorium. As a new method for preparation of actinide metals, the thermal dissociation of their intermetallic compounds with noble metals was developed. The actinide metals were further purified by repeated vacuum distillation and sublimation. High-purity protactinium metal was obtained by the VAN ARKEL-DE BOER procedure.

The intermetallic compounds were prepared by the synthesis of the elements (e.g. $AmPt_5$), by metallothermic reduction of the oxide ($CmBe_{13}$), or by hydrogen reduction of the oxide in the presence of noble metal powder ($CmIr_2$). Americium and curium pnictides were synthesized by reaction of group V metalloids (N, P, As, Sb, Bi) with actinide hydrides.

Americium metal (2g ^{241}Am) condensed from the vapour phase

Intermetallic compounds between beryllium and ^{241} Am, ^{238} Pu and ^{244} Cm, respectively,were produced as mechanically stable α-n-sources. Spontaneous fission neutron sources containing up to milligram amounts of californium oxide compacted in platinum have been fabricated.

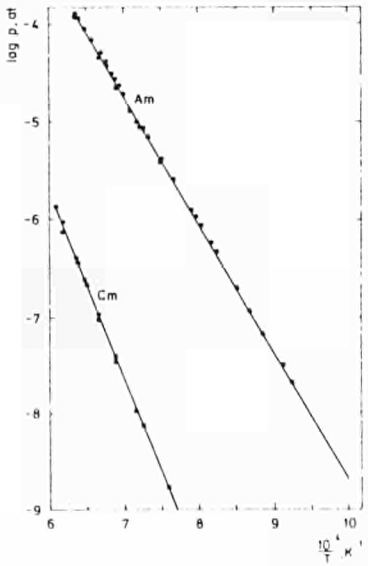
Chemical and Physical Properties of Actinide Metals and Compounds

The chemical and physical properties of solids depend on the number and the particular energy of the external electrons; their knowledge contributes to a better understanding of chemical bonding.

Crystal structure and lattice parameters, together with the results of chemical analysis, are used to characterize samples and to define reference states of the elements and their compounds to be studied. Although several of the first members of the actinide metal series crystallize in complex structures of low symmetry, americium and curium metal display the double hexagonal close packed structure which is also a feature of the light lanthanide metals. The radii and valencies of the metals are correlated. A new determination of the radius of actinium metal supports predictions based on relativistically calculated radii of the heavy metals; this is decisive for the assignment of electronic configurations and valencies to the actinide metals. The radii of transplutonium metals indicate valencies of three for americium and curium and show a tendency for divalency starting with californium.

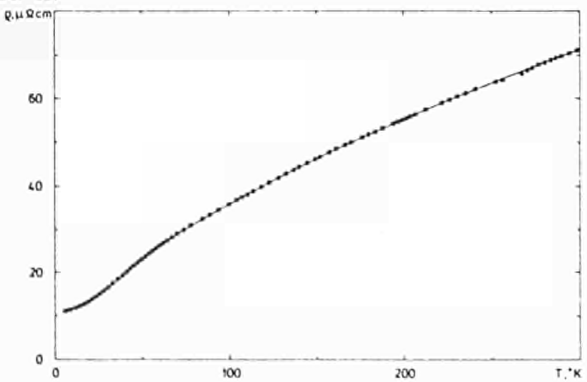
With well characterized metal samples, new values for the enthalpy of solution of americium and curium were obtained permitting the calculation of formation enthalpies of numerous americium and curium ions and compounds. The determination of the vapour

pressures of americium and curium by a KNUDSEN effusion technique combined with mass spectrometry furnished more precise values for the enthalpy of sublimation of these metals. These fundamental thermochemical quantities are required for the knowledge of the systematics of thermodynamics and the electron configuration of the actinide metals.

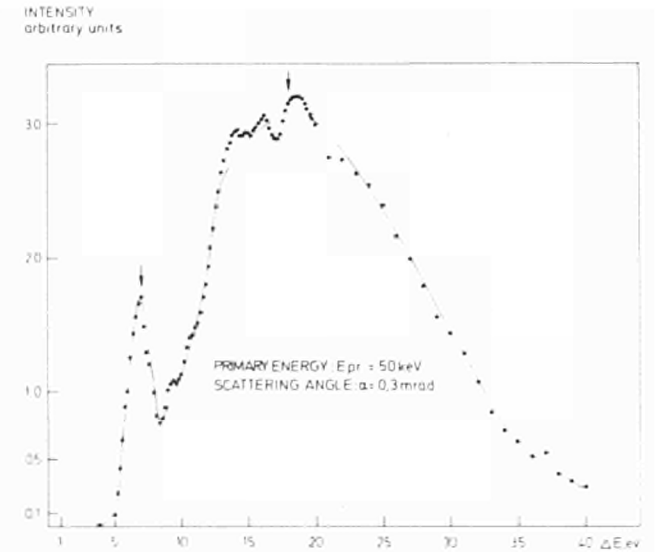


Vapour pressures of americium and curium

The magnetic susceptibility of americium and curium metals and of some of their compounds have been studied between room temperature and liquid helium temperature. Although the magnetic susceptibility of americium metal is almost temperature independent, the paramagnetism of curium follows a CURIE-WEISS law with antiferromagnetic ordering around 52 K.



The electrical resistivity of an Am-metal film



Electron energy loss spectrum of UO_2

Studies of the visible and ultraviolet optical spectra of the actinide elements and compounds give direct information on their electronic structure. A more detailed insight into the electron energy distribution is offered by the study of photoemission spectra. The first electron energy loss spectra of UO_2 were obtained. Infrared spectroscopy on oxides and on chalcogenides gives information about the bonding forces between actinide and metalloid atoms.

Transport properties such as thermal and electrical conductivities also depend on the electronic structure of the solid; at low temperatures the influence of radiation damage must be taken into account.

The study of the chemical bond in actinide solids begins to confirm the fundamental difference between actinides and lanthanides. Both series of elements are characterized by the presence of f-electrons. Although the 4f-electrons are well localized in the lanthanides, the 5f electrons of the first actinides are delocalized and able to participate in chemical bonding. Localization of 5f electrons seems to start with transplutonium elements; the last members of the actinide series show a marked tendency to lower valencies.

Some of these studies were carried out together with visiting scientists or groups of specialists working at various laboratories in the European Community and in the United States of America.

Selected Bibliography

The results of the work performed at the Institute for Transuranium Elements during the past ten years were published in more than 700 contributions to scientific and technical journals, to meetings and conferences, and in the form of external and internal reports. A condensed bibliography is given on the following pages.
Reprints and additional information on specific subjects can be obtained either directly from the authors or from the Laboratory's Documentation Service.

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