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COMMUNITY RESEARCH ON TRANSURANIUM ELEMENTS

THE WORK OF THE JOINT RESEARCH CENTRE OF THE EUROPEAN COMMISSION

Why research on transuranium elements?

Most of today's nuclear reactors are fuelled with uranium. Uranium is the only natural element capable of sustaining the nuclear chain reaction, that is, the process by which atomic nuclei split and release energy ("nuclear" energy), at the same time causing other nuclei to split and assure the propagation of the reaction.

But only a very small fraction (0.7 %) of natural uranium is useful for this purpose. The rest of it does not, because of its different nuclear structure, participate in the chain reaction. However, another important reaction takes place in this part of uranium as the result of the bombardment it is exposed to by neutrons coming from splitting nuclei: a gradually increasing number of the non-splitting uranium atoms are thus transformed into the atoms of another, new, artificial element: plutonium.

Plutonium has a very important feature: exactly like the 0.7 % of natural uranium, it can start and sustain the energy generating nuclear chain reaction. In other words, it is a "fissile" material. This means that the 99.3 % non-fissile natural uranium is not energetically useless; on the contrary, it can be regarded as a huge reservoir of potential energy, which can be tapped by converting it into fissile plutonium.

Plutonium is a transuranium element. It is so called because its place on the table of elements is beyond that of uranium, which is the heaviest element existing in nature. Uranium was therefore the last element listed on the table before the discovery of neptunium and plutonium.

Plutonium and other transuranium elements are continuously produced in ever increasing amounts in operating nuclear reactors. Most of them belong to the group of 14 elements called "actinides" because of certain atomic properties which are common to the elements following Actinium in the periodic table. This is why the research on transuranium elements is also indicated as research on plutonium and the other actinides.

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Research on transuranium elements is essential to the development of fuels for the fast breeder reactors of the future. Today's oxide fuels may have to be replaced by carbides of mixed uranium and plutonium with varying nitrogen content. However, while the best utilization of plutonium as a fuel is for fast breeder reactors (with their capability of producing more fresh plutonium in the uranium blanket than is burnt in the core of the reactor) it can also be put to use in today's commercial power reactors: here plutonium can either be burnt within the same uranium fuel elements where it is generated (thus helping to increase the life-time of the fuel element, or be recovered from spent fuel and recycled in new, plutonium-enriched fuel.

But what is just the subject-matter of research on plutonium and the actinides?

Apart from the solution of the numerous scientific and technological problems standing on the way of the development of economically and technically satisfactory plutonium fuel, major safety problems stem from the alpha-radioactivity and the toxicity of plutonium and the actinides.

Therefore, the acquisition of a full knowledge of the conditions for the safe handling of increasing quantities of transuranium elements, throughout the fuel cycle and up to the final disposal of radioactive wastes, is a "must" if nuclear power is to have any future at all.

These two faces of the same problem - technological development, and human and environmental protection - are therefore the object of research on transuranium elements.

Why centralized Community research?

Research on transuranium elements can only be conducted by highly trained personnel in very specialized facilities. Community action in this field is therefore prompted by economy and urgency. Economy in sharing out among member countries the burden of development costs and the inevitable risk of partial failures; urgency, because the development of economic, reliable and safe nuclear power is an urgent matter for the European Community.

Even at times of financial restrictions for public research budgets, the Community effort in the field of transuranium research is fully justified, especially if the amount of foreseeable future investments for energy sources is considered. On the basis of even the most conservative estimates, such investments from now until the end of the century will be of the order of many hundreds of milliards of dollars.

The European Institute for Transuranium Elements of the JRC

The Commission of the European Community has concentrated its efforts in the field of transuranium research by setting up the European Institute for Transuranium Elements at Karlsruhe, Germany. The Treaty between the Euratom Commission and Germany for the construction and operation of the Institute on the site of the German Nuclear Research Centre near Karlsruhe was signed on December 21, 1960, within the framework of the 1957 Treaty of Rome which established the European Atomic Energy Community (Euratom). The Institute is one of the four Establishments of the Commission Joint Research Centre (the three other are at Ispra, Italy; Geel, Belgium; and Petten, the Netherlands).

The Institute became fully operative in the course of 1967. It has special facilities for experimentation with all sorts of non-irradiated and irradiated uranium and pure transuranium elements, reactor fuels composed of them (ceramic materials made of mixed uranium and plutonium oxides, carbides, carbonitrides, etc.) and fuel rods where the fuel, under the form of dense pellets on vibrated powder, is clad by stainless steel tubes).

Cost and management of the JRC research programme for transuranium elements

Under the current 1977-1980 programme of the Joint Research Centre (JRC) transuranium research is conducted within the "Plutonium fuels and actinide research programm". The effort is kept at approximately the same quantitative level as that carried out during the previous, 1933-1976 programme. The 121 research staff responsible for the plutonium programme are assisted by 88 other members of staff. This personnel is essentially located at Karlsruhe, with about 5 % of them being located at Ispra, in order to make use of the expertise acquired there and useful to the plutonium programme.

The total budget for the 4-year programme amounts to approximately 41 million units of account.

The Director of TUI, in close consultation with the Advisory Committee for Programme Management (which is formed by national experts appointed by member countries), is responsible for the internal structure of the Institute and for the carrying out of the programme.

The procedure for programme preparation and adoption is initiated by a draft programme proposal by the Institute to the JRC Director General who, after consultation with the JRC General Consultative Committee (composed of industry, research and public administration representatives appointed by the Governments of member States), submits the proposal to the European Commission. This in turn, after its own review of the programme, conveys it to the Council of Ministers for final approval.

The scope of JRC research on transuranium elements

The JRC plutonium programme is to a large extent devoted to medium-term basic technological research. Part of it, on the other hand, concentrates on long-term fundamental research. The work so far carried out under the programme can therefore be divided into two main categories:

- Fuel science and technology involving studies of fuel fabrication techniques, investigation of physical and thermodynamit properties of plutonium based fuels, the preparation and execution of irradiation experiments and post irradiation structural and isotope analysis.

- Actinide research in which JRC is playing a central role within the Community both by contributing its own experimental and theoretical work and by coordinating an increasing number of cooperative research efforts from laboratories in various parts of the world.

The main aim of the fuel science and technology program has been to explore the limits of application of plutonium bearing fuels in nuclear reactors. These fuels are oxides, carbides, nitrides and carbonitrides. Of particular interest is the extent to which the lifetime of a fuel pin may be limited by volume increases of the fuel during irradiation (swelling). A better understanding of this phenomenon is of particular concern for the more advanced - non-oxide - fuels. Another limitation is presented by the chemical interaction between fuel and cladding material at elevated burn-up and in this case the main interest is in oxide fuel pins. In both instances important parameters which are not sufficiently well known are fuel temperatures, their distribution and evolution with time. These fuel science and technological studies have been complemented by detailed investigations of the - hitherto relatively unknown - production rate of fission products in a given neutron flux environment, and an extension of previous vapour pressure studies to extreme temperatures (at and above 5000 K) which are of interest for reactor safety studies.

The main aim of the actinide research, which has been carried out by JRC for the past four years, has been to contribute to the understanding of chemical bonding in actinide metals and compounds. Due to their particular electronic structure, the nature of the chemical bond in these materials gives rise to a number of unusual features in certain thermodynamic and physical properties. To further these studies it has been necessary to produce, separate and purify actinide elements and prepare well characterised samples of metals, alloys and compounds; and to perform physical and thermodynamic property measurements and investigate the possibility of relating the observations to the solid state electronic configuration of these materials.

<u>Current Status of JRC Research on Transuranium Elements</u>

A carefully planned research program has been carried out in order to identify the mechanisms contributing to the swelling of advanced fuels. The swelling is due in part to the precipitation of gaseous fission products and in part to the mechanical interaction between the fuel and its cladding. Laboratory experiments have been performed in order to define fuel properties (mainly mechanical and thermal) of interest for swelling. Fast reactor irradiations were carried out to burn-ups of 1 to 8 % in helium-bonded pins, and 1 to 4 % in sodium-bonded pins, with linear heat ratings up to 1350 W/cm. The dimensional and structural changes of the pins have been analysed. Models describing the in-pile behaviour of the test pins, in particular their volumetric changes during irradiation, have been designed.

As a result of these efforts it has been possible to build up a relatively complete semiquantitative picture of fission gas release and swelling in advanced fuels. For given initial and operating conditions, it is possible to define the operational limit.

Extensive investigations of the processes leading to cladding corrosion under irradiation have been carried out with a view to finding appropriate means for keeping these processes under control.

As a result some basic aspects of corrosion phenomena have been clarified by sophisticated analytical methods. Post irradiation examination of fast reactor fuel pins revealed how cladding corrosion depends on important initial and operational parameters. Corrosion simulation experiments revealed that some elements penetrated into the cladding much deeper than the visible corrosion layer. For the first time, the oxygen potential of milligram amounts of mixed oxide was determined via EMF measurements with a microelectrode. Cesium distribution in steel was measured by Rutherford back-scattering and gamma spectrometry.

By combining these experimental findings with data and theoretical predictions from the literature, it has been possible for the first time to establish a hypothesis on the likely mechanism of cladding attack.

Knowledge of the thermal behaviour of fast reactor fuel pins has been improved by thermal conductivity measurements of unirradiated fuel samples with various chemical compositions and mathematical models for thermal fuel pin analysis have been improved.

To extend these studies, new methods for the in-pile measurement of fuel temperatures and fuel-to-cladding thermal contact conductance have been prepared. A new type of temperature sensing device, based on the measurement of the temperature dependent velocity of sound in an appropriate sensor, has been developed. A new method for gap conductance determinations was also tested in the laboratory and is being adapted for in-reactor operation.

Newly developed highly automated equipment has been used for isotope analysis of heavy nuclides and certain fission products during irradiation of fuel rods. The techniques are used for analysis of fuel subjected to a fast neutron environment. The accuracy of the data obtained from these measurement techniques is well within the more demanding requirements of present day nuclear technology. The results obtained have been applied to fuel management problems and procedures for supervising the flow of fissile materials through fabrication plants, reactors and reprocessing installations.

An entirely new technique has made it possible to measure high temperature thermodynamic properties of fuel materials at temperatures up to 5000° K. This information is particularly important in reactor safety studies. The first quantitative results were obtained for uranium oxide and mixed uranium-plutonium oxides in 1974. A special technique allowed later extension of these measurements to temperatures below 4000° K.

As a first step towards the future work on the high temperature thermodynamic properties of advanced fuels, the evaporation behavious of these fuels has been measured up to 2500° K.

Partly in collaboration with other laboratories, it has been possible to determine the crystal structures of metal phases of actinium, protactinium, americium und curium; to determine the high temperature properties of certain actinide metals; to determine the electrical properties of americium and curium; to investigate the magnetic properties of protactinium, americium und curium, and to study the optical properties of metals and oxide systems. These investigations have clarified the role of the solid state electronic configuration of actinides. Optical spectroscopy has recently been complemented by other spectroscopy techniques which yield additional information on how the actinides are built up.

The actinide research work is devoted more to the basic properties of the lesser known nuclides beyond plutonium. In this area it has been no small achievement since 1973 to have purified multi-gram amounts of americium, gram amounts of protactinium and curium and milligram samples of actinium and californium. These precious quantities of materials are produced in the form of metals and compounds and are used for a wide range of experimental measurements.

Planned activities

On the basis of the multiannual programme started on January 1, 1977, the activities described above will be continued and supplemented by research on the safe handling and other safety aspects of transuranium elements and the fuels produced of them. The present programme is accordingly divided into three main research sectors:

- Study of the utilization limits of plutonium bearing fuels: this study involves research on the swelling of advanced fuels submitted to extreme operating conditions from the point of view of power density and in-pile time; investigation of stainless stell cladding behaviour and interaction with oxide fuel; and study of thermal behaviour, and thermodynamic properties at extreme temperature (5000°).

- Study of plutonium and actinides in the fuel cycle, including research on the formation and behaviour of actinides in different fuels in a fast neutron environment; research on the handling problems of plutonium compounds in view of their toxicity; and research on some aspects of reprocessing techniques for irradiated advanced fuel pins.

- Finally, the research work on actinide elements will be continued. Samples of rare and highly reactive protactinium, americium and curium metals, and their compounds will be prepared partly by innovative methods at the Institute. They will be characterized by chemical, metallographic and crystallographic analysis. Furthermore, the properties which will be investigated are the structure and the stability of actinide crystals; the thermodynamic parameters, such as the amount of heat involved in the process of formation of these cystals, and the specific amount of heat required to vary by a given value the temperature of each material; and the electronic parameters such as electrical resistivity, and phtoelectron and optical emission. The investigation of these properties of actinides will be also performed in collaboration with specialized laboratories within the Community that have established expertise in actinide or actinide-related research areas.

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