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COMMISSION OF THE EUROPEAN COMMUNITIES

Multiannual Programme of the Joint Research Centre 1980—1983

1982 Annual Status Report

Plutonium Fuels and Actinide Programme

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Plutonium Fuels and Actinide Research 1982

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Projects:

1 Operation Limits of Plutonium Fuels
1.1 Swelling of Advanced Fuels
1.2 Oxide Fuel Transients
1.3 Equation of State of Nuclear Materials

2 Actinide Cycle Safety
2.1 Formation of Actinides (FACT)
2.2 Safe Handling of Plutonium Fuel (SHAPE)
2.3 Aspects of the Head-End Processing of Carbide Fuel (RECAEB)

3 Actinide Research
3.1 Crystal Chemistry
3.2 Solid State Studies
3.3 Applied Actinide Research

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Introduction

Nuclear energy is released by fission of heavy nuclei. In parallel with the fission process in a nuclear reactor, neutron capture produces elements heavier than the initial uranium. These so-called transuranium elements such as plutonium and americium have important consequences for the nuclear fuel cycle and, because of their high specific radioactivity, particularly the biologically important alpha particles which they release, must always be handled with care.

Technological aspects of the handling and behaviour of these substances during all stages of the fuel cycle, fabrication, irradiation, reprocessing and waste disposal, are thus of importance for the continued development of nuclear energy. All necessary information should be available on the physical and chemical properties, not only of uranium compounds but also of those of plutonium and higher actinides.

Nuclear safety programmes focus on the avoidance of accidents in nuclear installations and the evaluation and amelioration of the consequences should some unexpected failure lead to a dangerous situation. The source of the danger is the nuclear fuel which has to be contained so that neither fission products nor the highly active actinide elements can reach the environment.

Thus basic research on the technology of nuclear fuels, especially those containing plutonium still has an important place in nuclear energy. The field extends already beyond plutonium as breeding and recycling require a better knowledge of the nuclear and chemical characteristics of the higher actinide elements and their compounds.

The European Institute for Transuranium Elements was established with all of the necessary facilities for this work and it serves as the focal point for a Community Research Programme,

- meeting demands not satisfied in national laboratories
- giving all Member States immediate access to this area of research
- extending research on nuclear fuels in preparation for the future development of the cycle
- coordinating dispersed efforts in the field of basic research on actinide elements.

By a regular exchange of ideas, experience and plans

with national laboratories the Institute ensures that its activities are relevant and complementary to other programmes.

The strong emphasis in the Joint Research Centre Programme on Nuclear Safety has resulted in some reorientation of the work, studies of light water reactor fuel behaviour being introduced with a consequent reduction in activity on fast breeder fuels. An exception is work on the so-called "advanded fuels" which continues and which is now directed towards the specification of a fuel having optimum in-reactor performance.

Results

1. Operation Limits of Plutonium Fuels

1.1 Swelling of Advanced Fuels

The objective of this sub-project is to provide experimentally based models of the mechanisms determining the performance of fast reactor advanced fuels under the full range of normal operation conditions.

The general picture developed during the preceeding years for the whole range of fuel compositions (carbide, carbonitride, nitride) has been quantified mainly for carbides during 1982 in the following areas.

Fission gas swelling in carbide fuel

Fission gas swelling is the most important mechanism which determines the in-pile performance of an advanced fuel besides its mechanical properties. Especially in the outer dense zone of a fuel pellet where fission gas release is negligible or at least low, the gas distribution between various bubble populations and gas in solution and the associated swelling contributions have to be known. A systematic study has now been concluded by the analysis of the small fission gas bubbles with diameters in the range 3 to 20 nm by transmission electron microscopy. The larger bubbles with 30 to 400 nm diameter had been investigated in previous years by replica electron microscopy. The unexpected result is that up to 11 a/o burn-up 50 to 70% of the gas is kept in small clusters and/or dynamic solution in the fuel matrix and the small bubbles (<20 nm diameter) contain only 5 to 10% of the gas. Consequently their contribution to swelling is very small. Fig. 1 shows a transmission electron micrograph with bubbles ranging from 5 to 50 nm diameter.

In-pile performance of Na-bonded fuels

The above results were then used together with other swelling data evaluated previously to establish a complete swelling balance in Na-bonded carbides. The total swelling as function of burn-up can be represented by a simple formula. This is used to describe the useful burn-

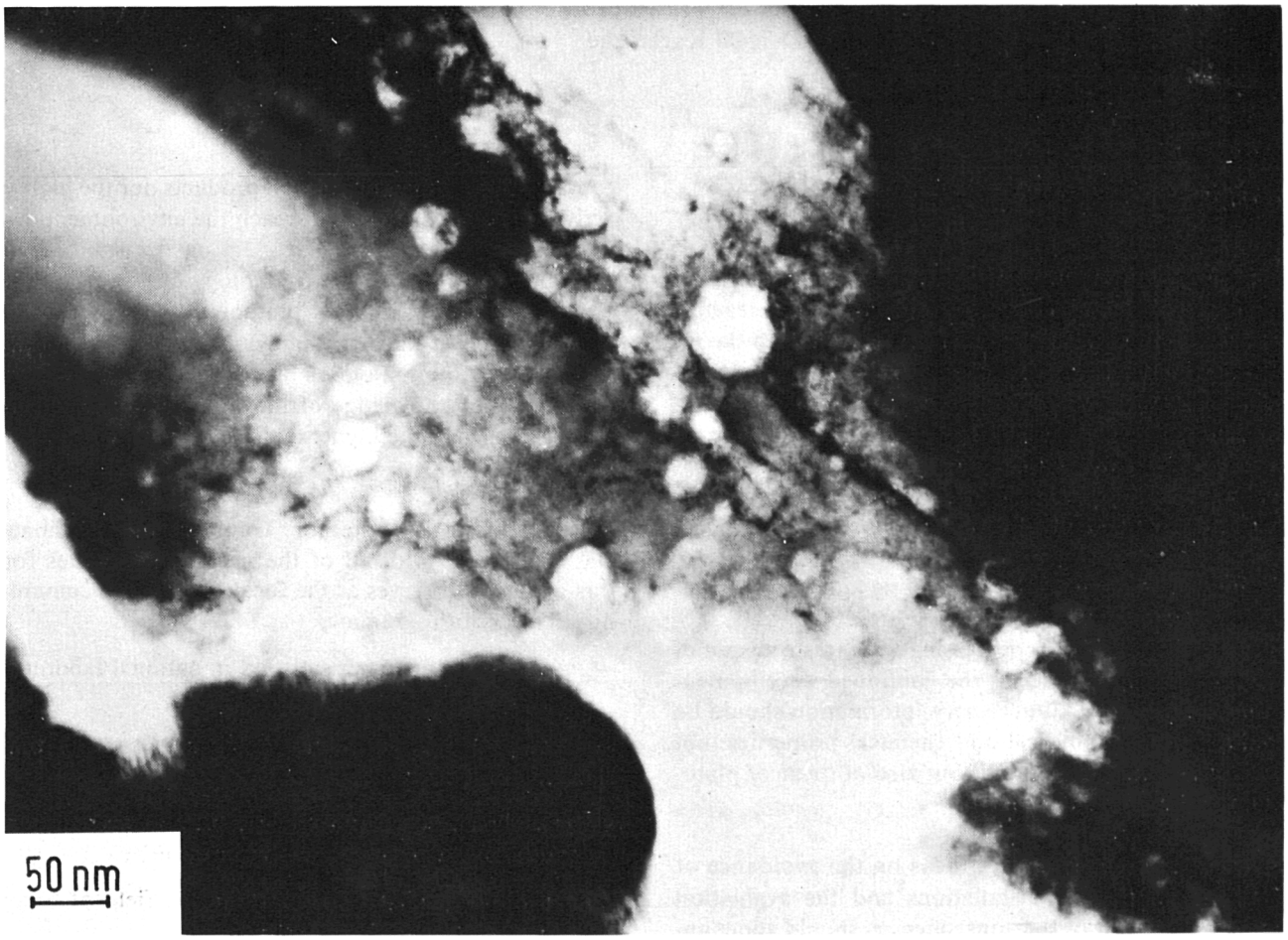

Figure 1 Small fission gas bubbles seen by transmission microscopy in a section of the sodium-bonded fuel pin CEA C2/4. Burn-up 8.9 at-%; temperature 900° C

up of a Na-bonded carbide pin as function of linear heat rating and of initial fuel/cladding gap, i.e. of the free space available for swelling. Furthermore the conditions for cladding carburisation under Na-bonding conditions were quantified and the differences in swelling performance between carbide, carbonitride and nitride could be introduced approximately into the swelling formula. Thus a rather complete description of the Na-bonding pin concept has been provided. Finally, problems were identified on which further fuel development should be concentrated if a fuel for the Na-bonding pin concept is to be optimized.

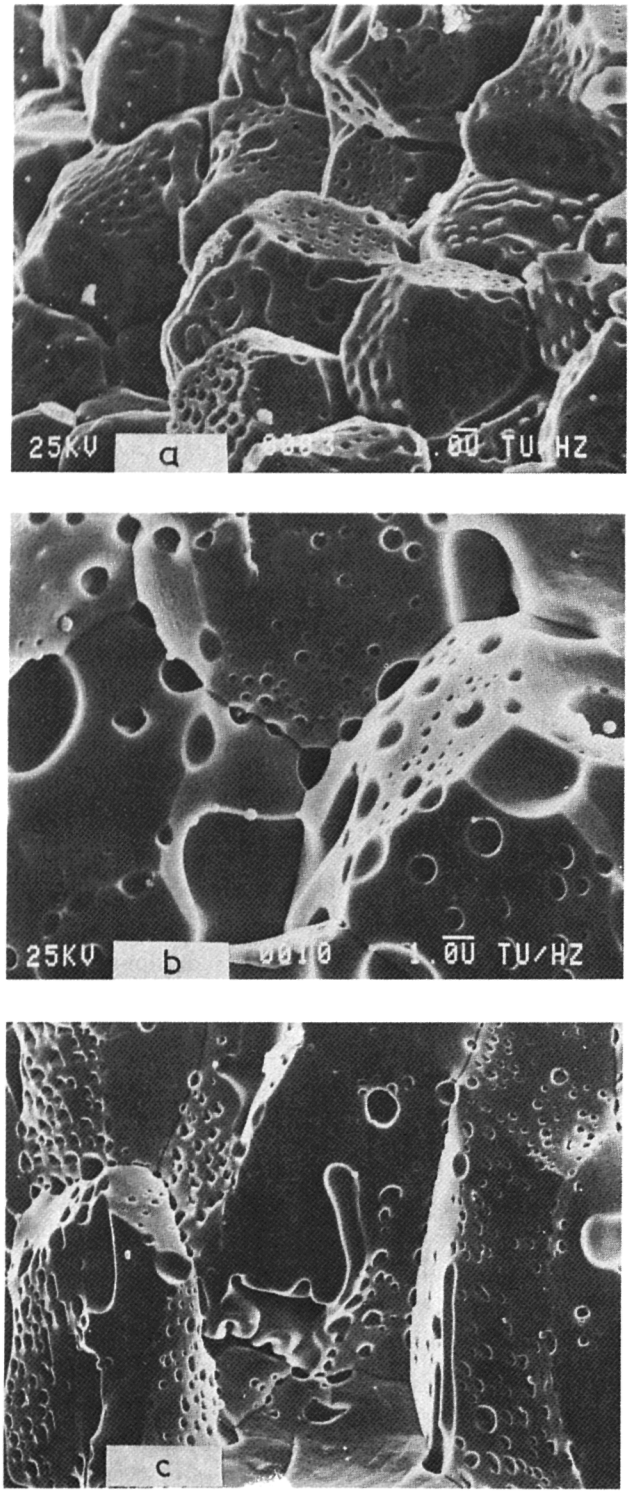

Fig. 2 Study of fission product precipitation at the grain boundaries in the first transient irradiation experiment TRANSON
a) near outer surface, transient temperature 2000° C
b) mean radius, transient temperature 2300° C
c) near centre hole, transient temperature 2700° C

Mechanical properties of advanced fuels
Previously systematic variations of the fracture stress in two-phase carbides, monocarbide + sesquicarbide, were established. These have now been analysed on the basis of structural fracture mechanics. This has opened a way for "tailoring" carbide fuels, i.e. for developing, within certain limits, fuel pellets with desired mechanical properties.

1.2 Oxide Fuel Transients

Transient conditions arise from operational events such as fuel changing, load changing or maintenance requirements, all of which may be classified as normal operation. Additionally unplanned transients will inevitably occur. Examples of such transients are equipment faults or operator errors which could give rise to mismatch between coolant flow and power generation leading to excursions which are limited by the reactor safety equipment. It is necessary to evaluate the consequence of any such transient on the fuel behaviour and its subsequent life.

It is the objective of sub-project "Oxide Fuel Transients" to develop on a theoretical and experimental basis a physical model of the fuel response during power and temperature transients from normal operation temperature to the melting point.

During the reporting period,

- examination of the first in-pile transient experiment has been completed and the next two experiments in the series have been irradiated. Fig. 2 illustrates fission product precipitation at the grain boundary following transient tests.
- fission product kinetics in fuel materials has been studied by implanting Te, I and Cs ions in UO_2 using a heavy ion accelerator. Release was measured following systematic annealing experiments to determine the temperature dependence of diffusion and to quantify the difference in release of the various Kr and Xe isotopes as function of their precursors.
- the effect of the fuel structure on fracture properties up to high strain rates has also been studied. This is the first stage of investigation of the effect of mechanical properties on fuel behaviour and fission gas release during transients.

— as a result of modelling work the code FUTURE has been modified to evaluate the effect of transients on a fuel pin. This is now being tested and compared with experimental observations.

1.3 Equation of State of Nuclear Materials

The equation of state of fast breeder fuels, and UO_2 as its main constituent, is required for safety risk assessments. The pressure contribution of the fuel is necessary up to at least 5000 K in order to estimate the energy release in a loss of flow (LOF) driven hypothetical core disruptive accident (HCDA) of a fast breeder reactor.

Various transient-type dynamic pulse heating techniques were developed to extend measurements up to 5000 K. On an average the measured total pressure above UO_2 turned out to be higher than the pressure calculated using thermodynamic extrapolations. At 5000 K the ratio of the measured to calculated pressure ranged from 2 to 10, depending on input data and the oxygen potential model used for the calculation.

The objective of the present efforts of the sub-project "Equation of State of Nuclear Materials" is to achieve consistency between the measured and calculated pressures above liquid UO_2 up to 5000 K, and to draw the necessary conclusions on the ternary oxide fuel $(U, Pu)O_2$. The critical analysis of the various thermodynamic extrapolations and investigation of thermionic emission were thus continued.

During the reporting period,

— Previous total pressure measurements, applying a laser pulse heating technique and depth evaluation, were confirmed by a specially designed collection technique, showing that complete elimination of the radial liquid displacement was achieved.

— Thermionic emission and its angular distribution were measured by various collection, deflection and probe diagnostics.

— A model for electron and ion emission from laser heated UO_2 surfaces, leading to the formation of a surface charge, has been developed for temperatures up to 5000 K. The additional and previously overlooked enhanced evaporation, caused by the thermionic emission at these extreme temperatures and the resulting surface charge, occuring under any conditions of free evaporation, allowed the discrepancy between measured and calculated pressures to be resolved.

— The Hertz-Knudsen-Langmuir equation which relates the rate of evaporation to the saturated vapour pressure has been generalised to account for thermionic emission at high temperature.

— A first experimental confirmation of the surface charge theory and the predicted cross over in sign of the potential corresponding to this surface charge was obtained.

— A special probe for flowing plasma has been designed to measure the charge density and the electron temperature in the laser produced gas jet.

— The various partial pressures over UO_2 were calculated at 2150 K, using a Schottky-Wagner intrinsic disorder model to determine the oxygen partial pressure, and then extrapolated to 5000 K accounting for the specific heat.

— Results were compared with the calculation of Green and Leibowitz (ANL) which is based on the Blackburn oxygen potential model, originally developed for non-stoichiometric uranium dioxide. At $UO_{2.00}$ this evaluation, assuming a thermal disorder of only three species U^{6+}, U^{4+} and U^{2+}, leads to too high oxygen partial pressures and thus to improbably large contributions in UO_3, UO_2 an O. The application of the Schottky-Wagner intrinsic disorder model overcame this difficulty.

— The perturbed hard sphere model has been applied to UO_2.

The direct measurement of the still remaining item of uncertainty in data extrapolation, i.e. the specific heat of liquid UO_2, is being prepared for temperatures up to 5000 K. A UO_2 droplet will be heated within a high pressure cell by a laser pulse, applying beam splitting under a tetrahedral configuration. The specific heat will then be evaluated from the cooling curve.

— The newly developed sub-microsecond six-wavelength pyrometer has been prepared and tested for this application.

— Various feasibilty studies on additional energy transfer by impact and thermal diffusion were completed.

In the same experimental set-up holography will be used to measure the thermal expansion of liquid UO_2.

— The holographic technique for expansion measurements of a small sphere (diameter of the order of 1 mm) in the laser autoclave has been successfully tested.

2. Actinide Cycle Safety

2.1 Formation of Actinides (FACT)

The objectives of this activity are

— to determine experimentally the nuclear properties of actinides irradiated in thermal and fast reactors in order to check the reliability of reactor physics calculations,

— to assess the irradiation behaviour of significant quantities of mixed uranium—minor actinide oxides.

During the reporting period,

— in spent PWR fuel (standard and MOX) the occurance of the less abundant uranium and plutonium isotopes ^{232}U, ^{236}Pu and of all curium isotopes (mass number 242 to 247) were experimentally determined for the first time and compared with ORIGEN predictions. Deviations of up to 100% were observed.

— to study the nuclear and material properties of minor actinides two irradiation experiments in KNK II were prepared in cooperation with KfK and CEN, Mol.

— bulk quantities of $^{241}AmO_2$ were separated from Pu fuel fabrication waste in cooperation with ALKEM.

— the test irradiation of an $(U_{0.5}Am_{0.5})O_2$ fuel in FR-2 was partially examined. The resonance shielding of ^{242}Am in a thermal neutron flux had been underestimated, which lead to higher burn up at the surface and ^{242}Cm formation. No significant fuel-clad-interaction was visible.

2.2 Safe Handling of Plutonium Fuel (SHAPE)

Handling of plutonium in nuclear fuel fabrication facilities leads to formation of aerosols which constitute a potential hazard to the operators. The acceptability of plutonium fuels in nuclear energy depends therefore also on the knowledge of aerosol formation, stability and distribution as a function of the type of fuel and on the availability of fuel fabrication procedures yielding reproducible and uniform products which meet all safety standards. The objective of the SHAPE activity is to study these aspects under realistic conditions.

During the reporting period,

— artificially generated mixed oxide, mixed carbide, mixed nitride and mixed carbonitride fuel aerosols were characterised by four different techniques: Inertial spectrometry, sedimentation counting, optical counting and scanning electron microscopic image analysis. Interrelations were established between the different "diameters" thus obtained,

— in aerosol samples taken from a plutonium fuel production chain, differences in characteristics were found, depending on whether the material involved was produced by mechanically blending uranium- and plutonium-oxide powders or via a sol-gel coprecipitation procedure.

— in glove box fire tests, contamination of glove box material was simulated with rare earth oxides, and first attempts were made to trace the spread of these oxides in an accident situation by neutron activation analysis,

— a new technique for producing mixed uranium-plutonium carbides by "direct pressing" was developed, which avoids one step of the conventional fabrication procedure, thus reducing the risk involved in handling the material.

— a study to investigate optimum conditions for obtaining homogeneous sinters from mechanically blended uranium- and plutonium powders at temperatures lower than those currently employed was brought to a successful end.

2.3 Aspects of the Head-End Processing of Carbide Fuel (RECARB)

Future reactor fuel, irrespective of the reactor type (LWR, HTGR, FBR) and of the fuel type (oxide, carbide) is expected to be reprocessed by TBP extraction after preparation of a nitric acid feed solution in a corresponding head-end process.

The objective of this activity is to study planned head-end processes for advanced fuel comprising:

— basic aspects of carbide fuel oxidation and dissolution,

— a comparison of different head-end reprocessing methods for advanced fuels,

— a test of a selected head-end process with an irradiated fuel pin.

During the reporting period,

— the technique previously developed to oxidise advanced fuels under CO_2 has been successfully applied to mixed carbides from the KfK Fast Breeder Programme (PSB) with a maximum burn-up of 8 at-%. Compared with the fuel from the DN-1 experiment the PSB material needed longer reaction times to complete the oxidation of the dense outer fuel zones,

— the composition of the dissolver off-gas has been measured by infrared absorption spectroscopy. The gas evolution curves obtained for the dissolution of (U, Pu)C are very similar to those from UC. The predominant gas during the dissolution is NO, whereas the evolution of NO_2 and CO_2 continues after the end of dissolution. The dissolution is faster with higher HNO_3 concentration and lower density of the material (Fig. 3),

— the photochemical process developed earlier for oxidation of the organic compounds present in the solution of dissolved carbides has been further applied to dissolver solutions. The oxidation is easier with irradiated material, due to the additional effects of the nuclear radiation (α, β, γ) on the cabonaceous by-products,

— small amounts of residues have been found upon dissolution of high burn-up MC ($\sim$6 at %) in

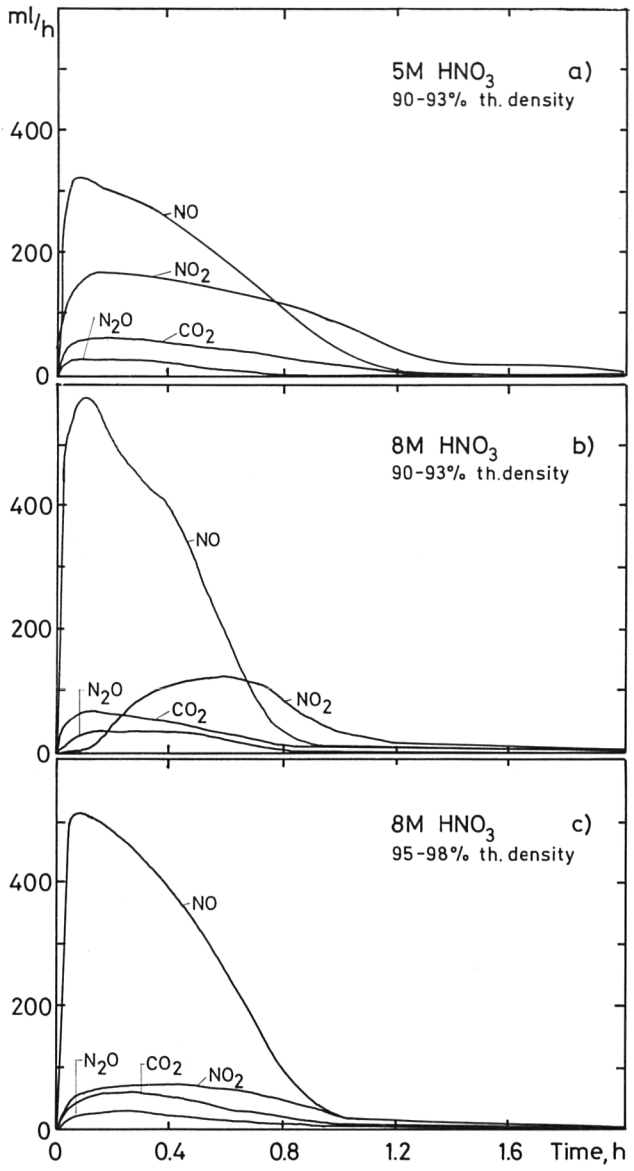

Fig.3 Gas evolution rate during and after dissolution of UC pellets in HNO_3 at 81° C

HNO_3, whereas unirradiated and low burn-up materials dissolved to >99.9%. The experimental conditions for dissolution of high burn-up MC can be chosen as to minimize the amount of residues. In 8 M HNO_3 at 80° C only 0.6—0.7% undissolved material was found (fission products included),

— uranium and HNO_3 were separated from simulated high activity waste ("HAW") solutions on a column containing TBP fixed on a polystyrene resin. This technique is useful for recovery of U and Pu from various fuel solutions and as a concomitant denitration step needed, if An-separations are to be made after the removal of U and Pu. Tests with a similar resin with HDEHP-Polystyrene as the extractant showed the TBP-Polystyrene to be superior in the above discussed aspects.

3. Actinide Research

The project emphasises the study of actinide metals, of intermetallics and of simple binary compounds (oxides, chalcogenides, pnictides, hydrides). The aim is to obtain information on the influence of 5f electrons and other outer electrons on different types of bonds (ionic, covalent, metallic) and to permit understanding and/or prediction of bonding related properties. This information is obtained by selected solid state investigations which require well characterized samples in a suitable form (e.g. single crystals, films etc.). Interpretation of the results depends on the development of bonding models on the basis of thermodynamics and solid state quantum physics in close cooperation between experimentalists and theoreticians.

The project is structured in the following activities.

3.1 Crystal Chemistry

This activity involves the preparation and characterisation of metals or compounds of known purity and structure, in a form suitable for the investigation of solid state properties. It is performed almost exclusively at the Karlsruhe Establishment.

During the reporting period,

— by recrystallization near the melting point in vacuum sealed tungsten crucibles single crystals of PuSb (Fig. 4) and PuAs have been grown, opening an entire new field of investigation. Magnetization studies (cooperation with ETH, Zürich (Switzerland)) have been performed and neutron diffraction measurements planned.

Also, single crystals of (doped) ThO_2 were grown by a high temperature solution growth method, using a $PbO\text{-}V_2O_5$ flux system.

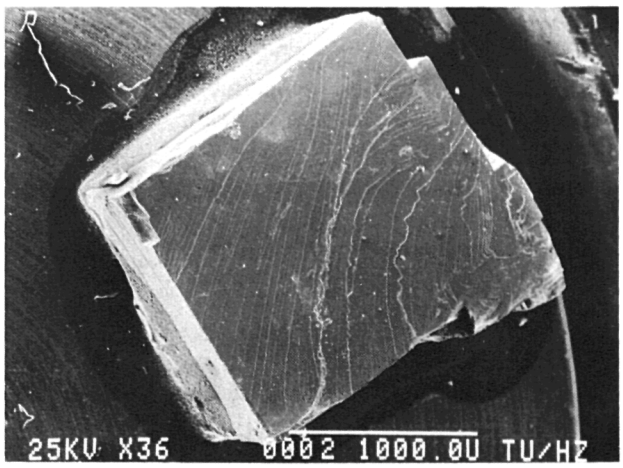

Fig.4 PuSb single crystal

3.2 Solid State Studies

Solid state properties are studied partially in collaboration with specialised laboratories within the community.

During the reporting period, at the Karlsruhe Establishment,

- by high pressure X-ray diffraction (HPXRD), the atomic volume and structure of Am and Cf have been studied under high pressure. This study is of great importance for the understanding of the 5f participation in the metal bonding of actinide metals (localization vs itineracy of 5f's),

  Also, by using synchrotron radiation from the DORIS storage ring (DESY, Hamburg) uranium monosulphide has been studied up to 40 GPa. A new tetragonal phase (US-III) has been observed, possibly a distortion of the NaCl-structure of uranium monosulphide in normal conditions (US-I).

- the low temperature structure of UCl_4 was determined by neutron diffraction. Also the structures of macrocyclic polyethers, possible complexing agents for actinide ions, were determined with the 4 circle X-ray diffractometer. Both studies are in the frame of structural research on strongly ionic actinide compounds.

- the thermodynamic and structural investigation of the (as yet unknown) protactinium-hydrogen system has led to the identification of several PaH_x phases and to the determination of the relevant thermodynamic quantities (enthalpy and entropy of formation).

- by photoemission (UPS-XPS), the **localization** of 5f electrons in americium metal and their **itineracy** in plutonium metal have been shown in a direct manner. This result is of extreme importance in actinide physics, since the itineracy/localization transition (Mott-like transition) between Pu and Am predicted by theory, had been up to now proved only through indirect evidence.

  Also, a study has been performed of the valence spectrum of plutonium metal in its α- and δ-modifications.

- the quantum theory of ground state properties of actinide compounds (e.g. cohesion) has been completed.

- the band structure and cohesive properties of thorium hydride (ThH_2) have been computed. A strong Th-H covalent bond, with almost no charge transfer accounts for the cohesive energy of this compound.

3.3 Applied Actinide Research

This activity offers support to other parts of the programme, when special developments — scientific or technological — in actinide-related research are requested on a short-term-basis. It includes preparation of encapsulated actinide samples for scientific research or as radiation sources, and the solution of problems of actinide recovery and waste fixation.

In the reporting period,

- it has been shown that radiation damage effects, as e.g. caused by curium inclusion, increase the mechanical stability of glasses and glass-ceramics which are possible waste carriers.

Conclusions

The programme of the Transuranium Institute has long included work on the so called advanced fuels for fast breeder reactors. Study of the swelling of carbide and nitride fuels is now nearing completion, the retention of fission gases in bubbles of different sizes in the fuel having been quantified as function of burn-up and temperature. The final step will be to define an "optimum" advanced fuel from the viewpoint of swelling behaviour under irradiation and a limited number of further irradiations is planned to test such fuels. Oxide fuel transient tests have begun and the model describing the behaviour of the fission products in the fuel pellet and swelling and release under a wide range of temperature transients has been prepared, this will continue to be developed and calibrated against the experiments.

An important step forward has been achieved in the studies of the Equation of State of Nuclear Fuels up to 5000 K. The discrepancy which had existed between theory and experiment in the vapour pressure over liquid UO_2 has now been explained.

Formation of some of the less abundant isotopes in PWR fuel has been determined experimentally for the first time and the first irradiation of a fuel containing equal proportions of uranium and americium has been examined giving important results required for the design of future experiments.

Aerosol formation during the fabrication of plutonium containing fuels, part of the activity "Safe Handling of Plutonium Fuel" has been studied using different sampling and analysis techniques. Characteristics of the aerosols generated using alternative fuel fabrication procedures have been compared.

Head-End Processing of carbide fuels has continued experiments with high burn up mixed carbides using both the oxidation by burning and dissolution techniques.

In the field of actinide research the preparation and characterisation of pure specimens is an important activity, subsequent property studies being carried out also in numerous specialist laboratories in European countries.

The protactinium-hydrogen system has been studied for the first time and an important confirmation by UPS-XPS of the localization of 5f electrons in americium metal and their itineracy in plutonium has been obtained.

A small but significant activity is the investigation of the effect of actinides on the properties of waste glasses, it has been shown that radiation damage can enhance the mechanical stability.

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