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REACTOR STRUCTURE MATERIALS: NUCLEAR FUEL

Background

In 1999, the fundamental nuclear fuel research was intensified in order to consolidate the scientific base for the applied research in this field. In the foregoing years, a growing need for basic materials research on nuclear fuel was felt. Only through a combination of both applied and fundamental research, the competencies to respond to rapid evolving demands in advanced research, can be developed. The reorganisation and concentration of several ongoing activities result in a more efficient use of the available resources. The fundamental research will focus on solid state research of nuclear fuel, on modelling of fuel behaviour and on the definition, technical preparation and execution of in-pile instrumented irradiation experiments. Applied research essentially remains market-driven, but with a strong incentive to develop activities that parallel more fundamental aspects and that have a long-term objective. From the year 1999, we will remember the organisation of a two day workshop on the "Impact of fuel chemistry on fission product behaviour" held June 16-17 at Mol, the set-up of a small thermochemistry laboratory, the finalisation of an important upgrade of the electron microprobe and a growing interest in spent fuel research.

Applied Nuclear Fuel Research

Contractual research on behalf of the international nuclear industry focussed in 1999 on the characterisation of fresh fuel and the effects of prolonged intermediate storage on spent nuclear fuel.

We developed an electron microprobe (EPMA) based methodology to analyse a polydispersed binary system and applied it successfully to several characterisation campaigns of un-irradiated MOX (on behalf of Belgonucléaire and COGEMA). The method has proven to provide excellent quantitative reliability in combination with a spatial resolution of 1μm.

The density of several batches of fresh MOX fuel pellets, as measured with a mercury pycnometer, confirmed within 0.2 % the density obtained on the basis of dimensional measurements of the pellets at the fabrication stage.

In the frame of spent fuel research, the influence of prolonged storage on the fuel morphology was evaluated. This year, we focussed on the evolution of spent MOX fuel after long term storage in ambient temperature and stored either under inert atmosphere or air. The elapsed time since end of life was 25 years for the analysed samples, and the fuel morphology aspects shortly after unloading and today were compared. Grain boundary alteration did occur in the air-exposed fuel samples (Fig. 1).

Fundamental Nuclear Fuel Research

Thermochemical fuel research

In 1998, we started a systematic investigation into the role of the grain boundaries in UO_2, particularly in relation to the transport mechanisms of fission products. Experimental studies of the chemical and

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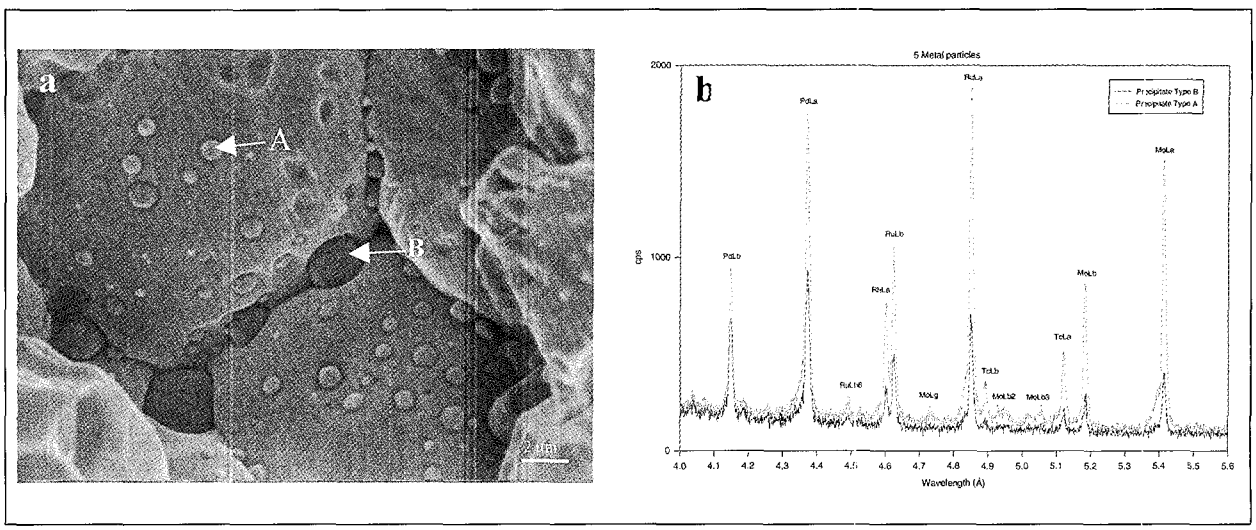


(a) Intergranular Decohesion Of Spent Fuel Sample Showing Large Metallic Precipitates On Grain Boundaries. Although The Morphology Of The Precipitates Is Different, Their Chemical Composition Seems To Be Identical (b).

physical characteristics of these grain boundaries were continued in 1999. The grain boundary characterisation in terms of uranium valence and uranate formation mechanisms was pursued further along the same lines.

The X-ray photoelectron spectroscopy (XPS) study of uranium valencies in bulk cesium uranates was concluded in 1999. This study showed the necessity for re-investigating the uranium 4f core level shift and peak broadening of simple uranium oxides (UO_{2+x}; $0 \leq x \leq 0.1$). Bulk samples of these uranium oxides with known stoichiometries were prepared in the furnace that was installed in 1999. The precise control of the gas atmosphere in the furnace allowed us to fix the stoichiometries. The subsequent XPS measurements have shown interesting effects on the peak parameters in the low hyperstoichiometries: a difference may be visible between the initial formation of oxygen defects (Willis clusters) and the subsequent crystallographic restructuring when higher oxides start to form.

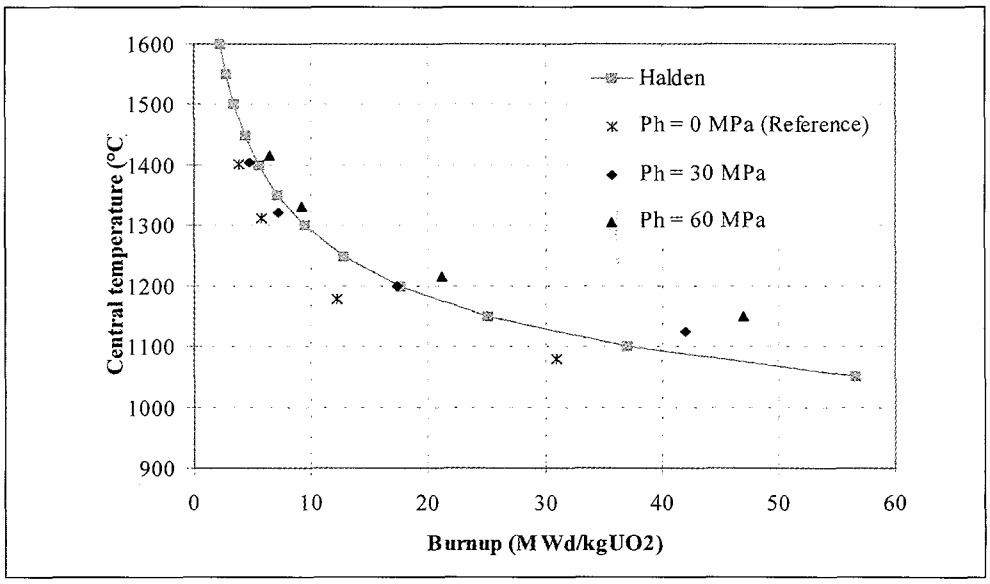
The system of the cesium uranates was further investigated to develop knowledge on the fundamental interplay of cesium and UO_2. These interactions were studied using thin layers of cesium deposited on UO_2 as well as films consisting of co-deposited uranium and cesium oxide. The possibility (found at the ITU (Institute for Trans-Uranium), Karlsruhe) to produce these samples in-situ (i.e. in UHV (ultra-high vacuum)) and subsequently analyse them with XPS without air exposure, provides us with clues on the grain boundary behaviour of cesium in irradiated fuel. This systematic comparison of the bulk and thin layer behaviour shall be continued in 2000.

Using the high-temperature furnace and gas atmosphere controller, we were able to produce UO_2 pellets containing reasonable amounts of cesium after sintering. Using these as model systems, we will try to deduce the mechanisms that govern cesium grain boundary migration in nuclear fuel by further XPS measurements.

The local chemical information provided by XPS turns out to be a vital piece of information still missing in the knowledge and understanding of the fuel - fission products system. The additional possibilities for model system engineering using the furnace system in combination with a future X-ray diffractometer with its proper possibilities for in-situ analysis, form a good start for building a more scientific understanding of the chemical and physical phenomena that govern the fission product behaviour in many different circumstances (i.e. in normal operating fuel, in incidental/accidental fuel and in spent fuel).

Modelling fission-gas release in Light Water Reactor (LWR) fuel

Since 1994 we are developing a new mechanistic model for fission gas release in LWR fuel. At present, the model embodies a large number of the underlying basic mechanisms of fission gas release and it couples the kinetics of the intra- and intergranular behaviour of the gas atoms. The microscopic module treats the behaviour of the fission gas atoms in spherical grains with a distribution of grain sizes. This module considers single atom diffusion, trapping and fission induced re-solution of gas atoms associated



The effect of the hydrostatic pressure on the central temperature for 1% average fission gas release versus burnup during irradiation at constant linear heat rate.

with intragranular bubbles, and re-solution from the grain boundary into a few layers adjacent to the grain face. The macroscopic module considers the transport of the fission gas atoms along the grain boundaries. Three mechanisms are incorporated as well: diffusion controlled precipitation of gas atoms into bubbles, re-solution of gas atoms into the adjacent grains and gas flow through open porosity when grain boundary bubbles are interconnected. The interconnection of the intergranular bubbles is affected both by the fraction of the grain face occupied by the cavities and by the balance between the bubble internal pressure and the local hydrostatic stress component.

The effect of the hydrostatic pressure on the central temperature for 1% average fission gas release versus burnup during irradiation at constant linear heat rate.

The model is under validation. To this end, the fission gas release model has first been coupled with the FTEMP2 code of the Halden Reactor Project for the temperature distribution in the pellets. We have simulated the empirical Halden criterion for fission gas release, relating fuel central temperature to burnup at which 1 % release can be exceeded. The general tendency of the fission gas release model is satisfactory

Integral experiments

In 1999, the fundamental decision on installing a refabrication/instrumentation facility was taken. The final goal will be to be capable to segment commercial length fuel rods of 4.5m into smaller segments and to equip these segments with thermocouple and pressure transducers. In this way, the non-instrumented base irradiation can be performed under rigorously representative conditions in power reactors avoiding lengthy (and costly) base irradiation campaigns in a materials testing reactor. The final instrumented test irradiation can then be performed in the materials test reactor taking full advantage of its capacity to perform experimental irradiation experiments of great variety and flexibility. In 1999, the laboratory made a first and important step in the realisation of a refabrication capacity in hot-cell. A compact, remote controllable and operable TIG welding set-up was designed and realised. Testing of the equipment will continue in the first half of the year 2000 and it will later be installed in hot-cell. Furthermore, SCK•CEN and the Halden Reactor Project formally agreed to realise the instrumentation equipment in our hot-cells.

Scientific output

Presentations

A. GYS, M. VERWERFT, MOCO I programme "Ceramographic observations and EPMA analysis of unirradiated MOX from Dessel – II. Ceramographic observations", R-3322, March 1999.

P. VAN UFFELEN, "Assessing the contribution of grain boundary diffusion to fission gas release in nuclear fuel", Enlarged Halden Programme Group meeting, Loen (Norway), HPR-351, May 1999.

S. VAN DEN BERGHE, "Concerto Programme: EPMA results, Electron Probe Micro-Analysis (EPMA) of Cr2O3-doped UO2", R-3336, May 1999.

A. LEENAERS AND M. VERWERFT, "Electron Microprobe Analysis (EPMA) of MOX productions", R-3360, June 1999.

P. VAN UFFELEN AND S. VAN DEN BERGHE edited the Proceedings of the Workshop "Impact of fuel chemistry on fission product behaviour", SCK•CEN report BLG-813, June 1999, with four independent contributions of the present group (from L. Sannen, S. Van den Berghe, P. Van Uffelen and M. Verwerft)

Contactual Reports

A. LEENAERS AND S. VAN DEN BERGHE, "Concerto Programme – Electron Probe Micro-Analysis (EPMA) of Cr2O3-doped UO2", R-3390, September 1999.

L. SANNEN, A. GYS EN M. VERWERFT, "Quality Assurance in Nuclear Fuel Research at the Laboratory of High- and Medium-level Activity at SCK•CEN", European Working Group on Hot Laboratories and Remote Handling, Plenary Meeting, Karlsruhe, Germany, October 13-15, BLG-831, October 1999.

P. VAN UFFELEN, "Modelling isothermal fission gas release", IAEA Technical Committee Meeting on technical and economic limits to fuel burnup extension, San Carlos de Bariloche, Argentina, November 15-19, 1999

M. VERWERFT, A. LEENAERS, M. LIPPENS AND L. MERTENS, "On the thermal evolution of Pu-rich agglomerates in MOX", International symposium on MOX fuel cycle technologies for medium and long term deployment, Vienna, 1999.

Reports

A. LEENAERS, M. VERWERFT, "DRYSTOR – Ceramographic observations and Scanning Electron Microscopy of long term stored fuel", R-3414, November 1999.

B. Vos, A. Gys, "POTOMAC – Final Report", R-3404, January 2000.

B. Vos, M. Verwerft, MOCO II programme "EPMA analysis and ceramographic observations on 3 non-irradiated MOX samples – I. EPMA analysis", R-3419, January 2000.

A. Gys, M. Verwerft, MOCO II programme "EPMA analysis and ceramographic observations on 3 non-irradiated MOX samples – II. Ceramographic observations", R-3420, February 2000.

Workshops

P. Van Uffelen and S. Van den Berghe organised the workshop "Impact of fuel chemistry on fission product behaviour", Mol, June 16-17, 1999.

L. Sannen and M. Verwerft organised a workshop on the "Nuclear Fuel Performance Research at the SCK•CEN Hot Laboratories", at the ABEN (Associaçao Brasiliera de Energia Nuclear), Belo Horizonte, Brasil, September 8-10, 1999.