

# ENRESA's R&D Programs on irradiated fuel and separation

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## Abstract

Enresa, the Spanish Radioactive Waste Management Agency was set up in Spain in 1984 as a state-owned company, in order to be responsible for management of this type of wastes in the country.

The company is supervised by the Government, to whom is obliged to submit an Annual Report of Activities and a Proposal for general Radioactive Waste plan on a yearly basis.

During the assumed 40-year lifetime of Spanish nuclear power plants the nuclear electricity production will generate 6800 tU of irradiated fuel and 84 canisters of vitrified HLW from reprocessing Vandellos I NPP(GCR) spent fuel (the reactor currently is in dismantling process). Also it is responsible of the following points:

- ◆ Management of low, intermediate and high level radioactive wastes (spent nuclear fuel)
- ◆ Operation of a facility for low and intermediate level waste disposal in El Cabril (Cordoba)
- ◆ Develop a program for selection and construction of a repository (spent nuclear fuel)
- ◆ Dismantling of nuclear installations

After creation of Enresa (1985) there was a change of strategy for HLW, LWR spent fuel is considered a waste. In different radioactive waste plans approved by the Government and in R&D programmes geological disposal is the only option.

In 98/1/29 the government takes decision of delay up to 2010 the approval by the Parliament of the option more convenient for final disposal according with the state of the technology.

The Research & Development programmes of Enresa linked to the general radioactive waste management plans are based in: Financing and management by Enresa. Development and creation of experimental research external groups (Universities, companies, institutions, ...). Closed linked to EU and international RDT programmes.

From 1986 up to 1999 ENRESA has implemented 3 R&D programmes. The principal objective of those plans was the development of technologies for the final waste management based in a deep geological disposal (AGP). Those plans were closely linked to the 3<sup>rd</sup> and 4<sup>th</sup> EU RDT programmes.

From 1999 - 2002 ENRESA 4<sup>th</sup> R&D plan (99-2002) in force, linked to the 5<sup>th</sup> EU RDT programme, will have as principal objective to deepening in knowledge and applications and verifications of the technological infrastructure created in previous plans based in irradiated fuel as waste form for disposal. The principal management option will be the storage of spent fuel in a repository. Partitioning and transmutation as way of decreasing volume and radiotoxicity

of waste form will be studied, so that, any decision than might be taken around year 2010, will be based on any feasible strategy.

A disposal concept for spent nuclear fuel and high level waste based on a deep geological repository was defined. In the last years, a large range of activities has been devoted to the performance assessment of a generic repository, called Enresa 2000.

Spent fuel performance assessment requires evaluation of its long-term ability to isolate and immobilise individual radionuclides after groundwater contact and it is necessary to know the waste behaviour under repository conditions.

The evolution of irradiated fuel under interim storage or deep geological conditions and the oxidation (O/M) rate before the water access to the fuel surface, is a factor of great influence on enhancing dissolution.

Those studies have been undertaken by means of chemical analogues, real spent fuel and artificially aged irradiated fuel.

This work presents results on ENRESA's previous Research & Development plans, and the activities in progress carried out specially on irradiated fuel, in accordance with the 5<sup>th</sup> General Radioactive Waste Plan (GRWP) in force, focused specially in tests performed with real fuel in hot-cells of CIEMAT or in international collaboration with Institute of Transuranium elements of Karlsruhe, although the main part of the program is based on use of chemical analogues of spent fuel, such as SIMFUEL and UO<sub>2</sub>, due principally to its lower costs and than presently there are not hot cells facilities in operation for PIE fuel studies available in Spain. Besides, there is a collaboration agreement on spent fuel research with ITU, following the guidelines of the GRWP to promote international co-operation.

The 5<sup>th</sup> General Radioactive waste Plan means a change in the orientation of the R&D that Enresa has been performing up to date. On the performance of spent fuel, this plan will focus in clarifying questions remaining open from earlier programmes; in order to increase understanding and improving the performance of irradiated fuel as a waste form. Basic research and infrastructure of other spent fuel management strategies will be implemented. The principal management option will be the storage of spent fuel in a repository. But partitioning and transmutation will be studied so that any decision that might be taken around year 2010 will be based on having enough information on any possible strategy.

A general overview and significant conclusions of this programme are presented, focused on projects in the field of spent fuel leaching and the research groups and infrastructure created and developed.

Some resources will be dedicated to exploratory research on partitioning and transmutation as high-level waste management strategies, aiming at show that a given elementary process could be efficient in the separation of nuclear materials.

In this direction has been created two new groups in Ciemat for studies on hydrometallurgical and pyrometallurgical separation, joined to the European projects in this area.

**KEYWORDS:** ENRESA; R&D programme; post irradiation examination

## 1. Overview of R&D projects on irradiated Fuel

### 1.1. 1<sup>st</sup> R&D Plan (1987-1991)

#### 1.1.1. General objective

The objective on spent fuel (SF) was the creation and improvement of research infrastructure to study the source term and containers as principal components of waste form taking into account that the unique option for definitive management was deep geological disposal (open cycle). A methodology was developed for studying the UO<sub>2</sub> as chemical analogue of spent fuel in a repository sited in salt.

The main results of this Plan were the development of a research group in Universidad Politécnica de Cataluña (UPC). Static dissolution test were performed with UO<sub>2</sub> as SF analogue, in oxidant and reducing conditions in NaCl and MgCl<sub>2</sub> brines. It was observed that the dissolution of this analogue was strongly dependent of pH and redox conditions and in less grade of the ionic strength of brines.

### 1.1.2. International collaboration

ENRESA participated in two international projects as LOFT and PHEBUS on severe accidents. It was the beginning of the international cooperation.

## 1.2. 2<sup>nd</sup> R&D Plan (1991-1995)

### 1.2.1. General objective

The principal processes and characteristics considered in the near field with respect to the second plan were:

- ◆ Composition and inventory (UO<sub>2</sub>, fission products, activation products, actinides)
- ◆ Dissolution/alteration of UO<sub>2</sub> (dissolution mechanisms, redox conditions effect, pH effect, salinity and composition effect of leachants, SF physical state effect (such as flaws, cracks, distance between pellets and grain size)
- ◆ Mobility of products dissolved (radiolysis effect, physical and chemical conditions effect, formation of complex and colloids, hydrodynamics conditions effect of environmental media.
- ◆ Gas generation
- ◆ Processes modelling

The activities related with the fuel have been oriented to obtain basic information on the dissolution processes under conditions very close to the natural systems. The main studies carried out with unirradiated UO<sub>2</sub> were:

- ◆ Dissolution mechanisms and models of UO<sub>2</sub>
- ◆ Dissolution model
- ◆ UO<sub>2</sub> dissolution in magnesium and sodium brines
- ◆ Dissolution tests in batch
- ◆ Experiments with SIMFUEL and UO<sub>2</sub> under irradiation

The studies carried out with SF were:

- ◆ Theoretic analysis of radionuclides release
- ◆ The main results of this plan were:
- ◆ Creation of a group and laboratory in CIEMAT for SF characterization and dissolution in batch and sequential studies by means of SIMFUEL and unirradiated UO<sub>2</sub>
- ◆ Consolidation of the UPC group with flow/through tests and UO<sub>2</sub> and uraninites studies and
- ◆ Collaboration with ITU and FzK to obtain data on SF

### 1.2.2. International collaboration

Participation in the 3<sup>rd</sup> EU Framework program: 1<sup>st</sup> project on direct disposal of SF and collaboration with FzK. In this period started the collaboration with EU ITU JRC.

## 1.3. 3<sup>rd</sup> R&D Plan (1995-1999)

### 1.3.1. General objective

The principal objective of this Third Plan is connected with the support necessary to carry out the High Level Wastes Plan, which studies the integral management of those wastes.

The generic objectives of this plan were:

- ◆ Verify the instrumental methodologies developed for the characterization of the geologic barrier and complete the necessary developments.
- ◆ Verify the operability and the viability of the engineering barriers, under real conditions of scale, temperature and depth, in accordance with the early designs studied for three lithologies (clay, salt, granite)
- ◆ Obtain basic data about behaviour of IF, claddings and containers.
- ◆ Obtain basic data on mobilization/retention processes of actinides and fission products through the clay barrier and the geological barrier

- ◆ Obtain basic data on radionuclide behaviour in biosphere

The objectives and lines on IF were:

- ◆ Dissolution mechanisms: redox conditions effects, pH effect, chemical composition of leachants as a function of barrier materials, physical effect of SF as a function of burnup.
- ◆ Mobility of products dissolved (physicochemical conditions effect, effect of environmental thermodynamics conditions, complex and colloids formation, engineering barriers effect)
- ◆ Gas generation
- ◆ Modelling

### 1.3.2. International collaboration

Regarding to the 4<sup>th</sup> EU Framework Program, ENRESA participated in different projects, between them one on management and storage of radioactive wastes. "Source Term" and "SPA". In addition, ENRESA participated with other international organizations such as ITU (Karlsruhe), FzK and AEA (UK).

## 1.4. 4<sup>th</sup> R&D Plan (1999-2003)

### 1.4.1. General objective

The objectives and activities considered in the present Plan are to make a decisive contribution to the construction of the scientific and technological bases supporting future decision/making on the most adequate way of addressing the definitive management of high level wastes. Furthermore, this R&D Plan should provide all the technology not currently existing within a conventional or commercial framework and required for the management of low, intermediate and high level wastes, including dismantling, radiological protection or environmental intervention in the event of radioactive contamination.

The areas of research included in the present Plan have been selected with a view to ensuring better compliance with the new directives included in the 5<sup>th</sup> General Radioactive Waste Plan (GRWP) approved by the Government in July 1999, and are the followings:

- ◆ Basic technologies of waste characterization and behaviour, applicable to various fields of management undertaken by ENRESA.
- ◆ Separation and transmutation
- ◆ Disposal
- ◆ Performance and Safety assessment
- ◆ Support of facilities: low and intermediate level wastes, dismantling and radiological protection.

The principal objective in SF is the study of radiolytic effect on spent fuel corrosion ( $\alpha$ ,  $\beta$  and  $\gamma$ ) by means of alpha doped pellets dissolution studies, and external beta and gamma sources on SF matrix. The studies on real spent fuel were made in collaboration with ITU JRC, by means of a collaboration contract on SF research signed in October 2000. The research on SF in the past was based principally in studies of analogues of spent fuel, as: uraninites, SIMFUEL (UO<sub>2</sub> matrix with stable nuclides simulating radioactive isotopes), and natural UO<sub>2</sub>. The research on spent fuel was carried out by means of international collaboration.

The instantaneous release of RN on real spent fuel dissolution and finally the studies of alteration/dissolution of real spent fuel in dynamic tests for develop a model of behaviour of SF in repository conditions.

A research collaboration agreement was signed with ITU-JRC (KA). In July 2000 and a special contract on spent fuel research in October 2000, focused mainly in studies on real irradiated fuel. In Ciemat and UPC research continue to be based in studies on irradiated fuel analogues.

In separation and transmutation the principal objective is creation of research groups in Ciemat for separation and consolidation of the group In Transmutation., for development of the scientific and technological bases for all possible options and management techniques that will facilitate decision - making at that time regarding the best option to be adopted for the management of this type of wastes in Spain.

Up to the moment was created two groups in Ciemat for research on separation by hydrometallurgical and pyrometallurgical processes. The hydrometallurgical group has joined to the EU projects Calixpart and Partnew which objectives are separation of long life RN originated in the Purex process of the reprocessing plants, by extraction with calixarenes and malonamides respectively and create laboratories infrastructure for carry out this works. Extraction with borated compounds will be explored in near future. Collaboration Ciemat - UAM, by which UAM synthesised organic molecules adequate for the extraction processes, has been signed.

In Pyroprocessing the objective is study applications of pyrometallurgical processes to the separation of Ln and An, by molten salts (Chlorides) and electrochemical processes. Collaboration in EU project Pyrorep will help to gain experience in this field by participation in this project. An agreement Ciemat - UVA and Ciemat - CEA has been signed for collaboration in this field. An agreement for collaboration on Separation is in negotiation with JRC-ITU.

In the following point, results on PIE studies on real irradiated Spanish PWR rods, stored in dry for more than 20 years at different cooling times are presented

## 2. Research and Development Program on special fuel rods

This Program was proposed to demonstrate in a commercial PWR the reliability of improving fuel rod designs for high-power and high - burnup application and evaluate fuel integrity after more of twenty years of dry storage. All special test fuel rods utilized Zircaloy - 4 cladding. As part of the fuel improved design, some of the rods were fabricated with internal pressurization to minimize clad strain during lifetime. Many of these rods were fabricated with spiked (high) enrichments to achieve the desired extended burnup and high power levels expected in advanced PWR cores. The physical characteristics (burnup, enrichment, internal pressure, etc.) of the rods tested are showed in Table 1.

This paper summarizes the results of the Research and Development Program, which was performed between Westinghouse, Junta de Energía Nuclear and Unión Eléctrica, S.A., between 1968 and 1977. The results of the non-destructive and destructive examination program of dry-stored rods agreed between AEA Technology and ENRESA between 1997 and 1999 and the results from agreement between the UKAEA and Westinghouse Corporation. Subsequently, AEA Corporation on behalf of CRIEPI/NFI, have commissioned a program of further examination of fuel rods. Subsequently, Summit AEA Corporation (Japan), on behalf of CRIEPI/NFI, have commissioned a further examination programme on these rods, at AEA Technology Windscale.

Following irradiation the rods were transferred to JEN's (at present CIEMAT) postirradiation examination facilities in September 1976, where a program of non-destructive and destructive examination was carried out. The rods were stored, from that time, in a specially designed transport cask (see the following figure 1), in which each rod was contained in a stainless steel capsule in air until they were shipped to AEAT (UK) in 1990.

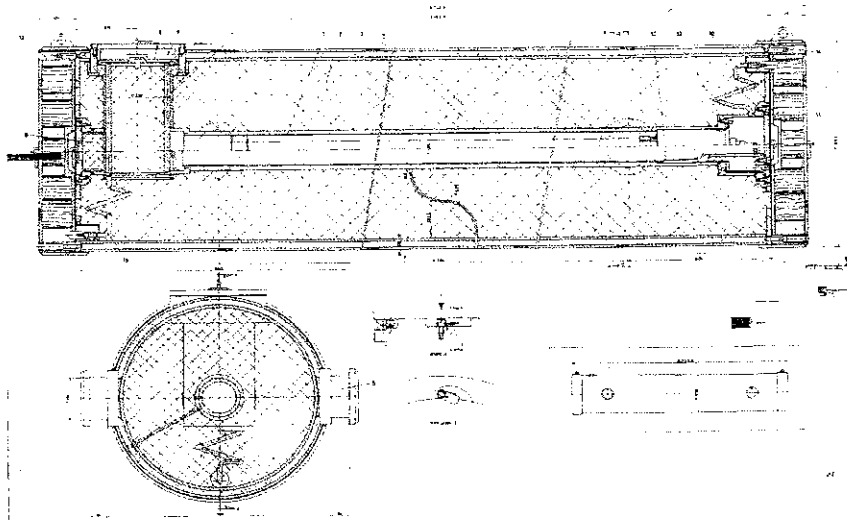


Figure 1 Transport cask.

Special test assemblies were irradiated in a PWR at different number of cycles, until a maximum of five cycles. There were two major design variables among the test rods: power level (as a function of fuel enrichment), which varied from 2.4 w/o to 6.60 w/o  $^{235}\text{U}$  (internal pressurization air at atmospheric pressure or He at 34.48 bar), and sintering temperature, which varied from 1492 to 1750 °C.

Table 1

Rod	Burnup MWd/kg U	Fuel enrichment $^{235}\text{U}$ (w%)	Nominal internal pressure bar (gas)	Sintering temperature °C
A	11.6			
B	11.5			
C	31.2	2.90 (Standard)	34.5 (He)	1560
D	49.6	4.14 (Spiked)	34.5 (He)	1550
E	57.2	3.60 (Standard)	1 (air)	1492
F	58.2	3.91 (Standard)	34.5 (He)	1492

## 2.1. Post irradiation Examination performed on fuel rods for PWR

### 2.1.1. Non destructive tests

#### 2.1.1.1. Visual appearance

The rods were examined for overall integrity, surface deposits, condition of the plug welds, grid contact marks, etc. Some of the rods were sampled for crud analysis. There were no significant anomalies, such as excessive corrosion at the end plug welds, fretting on grid locations, fuel rod bowing, plenum collapse, etc. The major surface feature on spiked enrichment (high power) fuel rods was local deposits of tenacious crud which could appeared first in Cycle 1 and did not change significantly in appearance during the next two cycles.

Rods A, B, C, D, E and F were covered in a thin base oxide layer. The intensity of this corrosion was seen to vary with the burnup of the rods, i.e. lighter on rods A and B ( $\approx 11$  MWd/kg U) but heavier on the higher burnup rods, in particular rod D ( $\approx 50$  MWd/kg U), which exhibited a much thicker, coalesced area rod corrosion. The typical appearance of rod D is shown in the following Figure 2.

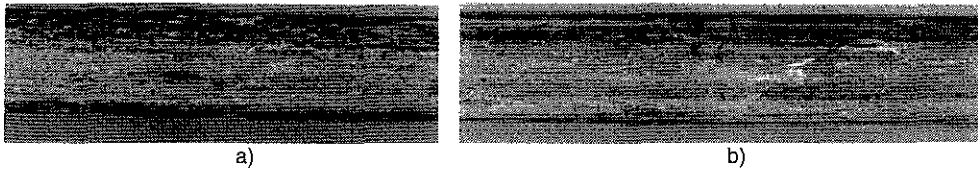


Figure 2 Typical appearance: a) rod A ( $0^\circ$ ), b) rod B ( $0^\circ$ ).

Nodular corrosion was slightly increased on all rods around the grid band region, and it is seen as lighter circumferential bands on the inspection tapes. See figure 2, which shows the appearance of rod B.

#### 2.1.1.2. Oxide thickness measurements

Cladding outer oxide thickness measurements were performed on each six rods sent to AEA Technology plc, using a Fischerscope Eddy T3-DO7 high frequency Induced currents probe, with a minimum test area of 3 mm on the rod outer surface. The measurements were made along 4 axial lines  $\approx 90^\circ$  apart, with continuous probe contact with the oxide surface. As an example the results from rod F is illustrated graphically in Figure 3. Initial measurements yielded data, which were considered to be very "noisy". This was attributed to dirt and crud picked up by the rods during transport, storage and handling. A decision was therefore made to lightly brush all six rods to remove this dirt and crud. The results of oxide thickness measurements are in the expected range for rods of these burnups, and are in good agreement with previous measurements on similar rods from the same reactor.

Figure 3 shows for comparison the "unbrushed" data obtained from rod F. The effect of brushing (compare Figure 4), aforementioned, is seen to be a reduction in the coating thickness of  $\approx 5 \mu\text{m}$ , and a significant reduction in noise level. The reduction, after cleaning, of about  $5 \mu\text{m}$  in the thickness layer suggests that there was a uniform covering of loose deposit on all of the rods, as well as any debris picked up during storage and handling.

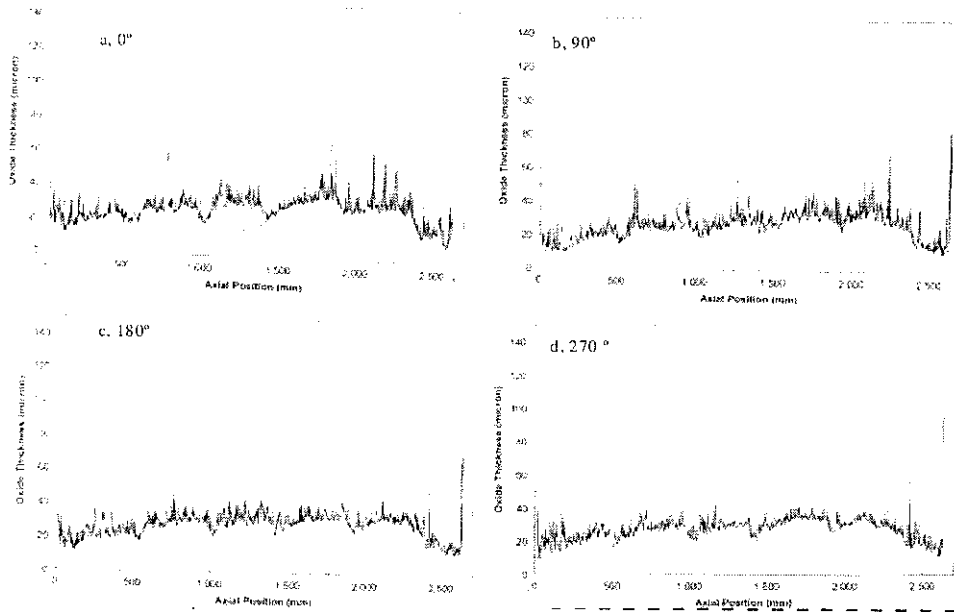


Figure 3 Unbrushed oxide thickness measurements from rod F ( $a = 0^\circ$ ,  $b = 90^\circ$ ,  $c = 180^\circ$  and  $d = 270^\circ$ )

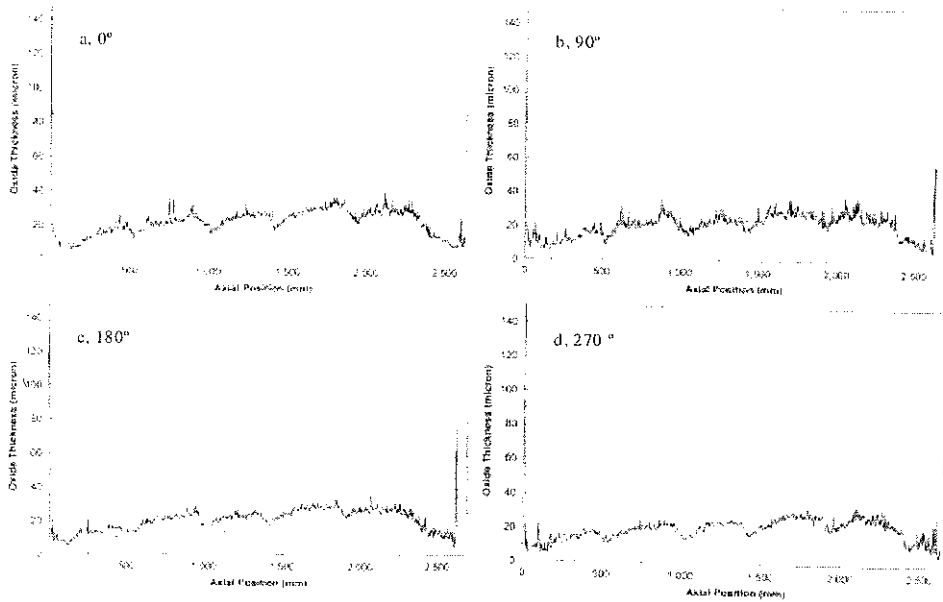


Figure 4 Oxide thickness measurements from rod F ( $a = 0^\circ$ ,  $b = 90^\circ$ ,  $c = 180^\circ$  and  $d = 270^\circ$ )



## 2.1.2. Destructive tests

### 2.1.2.1. Clad-fuel interaction

It should be noted that unpressurised high-power rods showed more extensive fuel-clad I.D. bonding at end of Cycle 2 than their pressurised sister rods. This is probably a consequence of greater clad creep down of the unpressurised rods at both firmer and longer contact with the fuel.

Electron images of the fuel-clad interface region corresponding to rod E are shown in Figure 5 for the 1091 mm sample position. The micrographs clearly show a bonding layer  $\approx 10 \mu\text{m}$  thick. Several small area ( $\approx 5 \times 5 \mu\text{m}$ ) analyses were made in this region for each sample. The results of these composition measurements are given in Table 2.

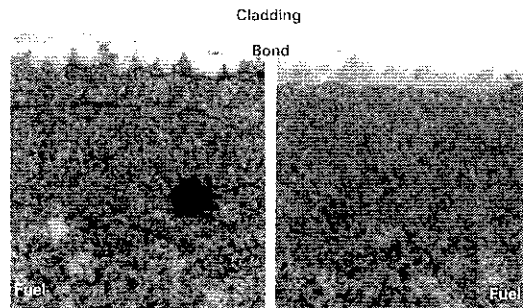
**Table 2** Elemental analysis of fuel cladding interface region (weight of percent, rod E).

Sample loc. (mm)	Analysis region	W/o												
		U	Pu	Xe	Cs	Zr	Mo	Ru	Ba	Ce	Nd	Te	I	O
1632	Cladding	0.000	0.075	0.000	0.000	98.445	0.011	0.000	0.000	0.263	0.103	0.012	0.000	1.090
	Bonding layer	3.513	0.223	0.572	0.592	71.038	0.420	0.297	0.452	0.656	0.303	0.072	0.000	21.861
	Bonding layer	0.071	0.052	0.170	0.007	85.785	0.207	0.079	0.082	0.292	0.161	0.008	0.000	13.087
	Bonding layer	0.613	0.100	0.554	0.202	74.851	0.3670	0.232	0.194	0.435	0.247	0.035	0.000	22.169
1091	Fuel	76.522	4.254	0.143	1.063	0.963	0.818	0.587	0.870	1.265	0.814	0.374	0.000	12.327
	Cladding	0.118	0.061	0.001	0.024	98.926	0.063	0.000	0.000	0.207	0.102	0.102	0.000	0.498
	Bonding cladding	1.487	0.287	0.519	0.152	74.331	0.548	0.450	0.240	0.480	0.408	0.408	0.000	21.034
	Bonding cladding	2.376	0.214	0.308	0.143	73.762	0.424	0.331	0.479	0.413	0.413	0.271	0.014	21.147
	Bonding cladding	1.754	0.000	0.416	0.155	74.602	0.406	0.317	0.220	0.429	0.429	0.306	0.000	21.337
	Fuel	74.600	3.602	0.231	0.873	3.133	0.948	0.831	0.871	0.227	1.227	1.088	0.031	12.200

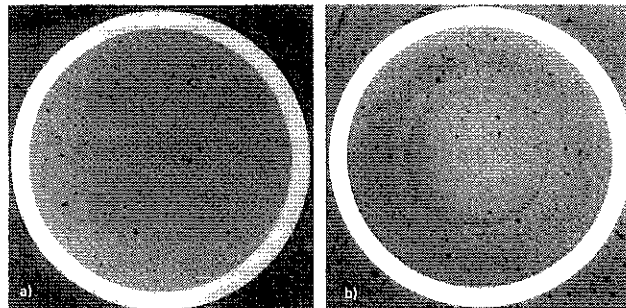
### 2.1.2.2. Metallography and ceramography studies

At low magnification, in rod E (after cycle 5), a central "core" became apparent, as shown by the macrograph in Figure 6. Further examination at higher magnification revealed a marked transition of microstructures across the fuel pellet radius:

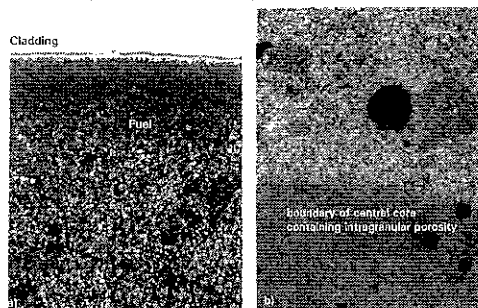
- ◆ At the pellet rim of rod E there is a very porous zone, where no grain boundaries could be discerned (Figure 6) the depth of this rim was in the range  $\approx 100 - 140 \mu\text{m}$ .
- ◆ Inwards from the rim of rod E to the edge of central zone (Figure 6) the fuel is very fine grained. The matrix had a mean linear intercept grain size of  $5 \mu\text{m}$ , with patches of even smaller grains ( $\approx 2 \mu\text{m}$ , Figure 7). In this zone there were very few intragranular features.
- From the cross section of the sample at 1632 mm of rod was observed a fast transition from inter to intragranular features at the edge of the central core. Again, in the central region the grain boundaries were difficult to etch up, although there appears to have been some equiaxed grain growth (by a factor of  $\approx X2$ , Figure 8).
- ◆ Overall there were many rounded macropores, generally in the size range 20 to  $> 100 \mu\text{m}$  in the samples of rod E.



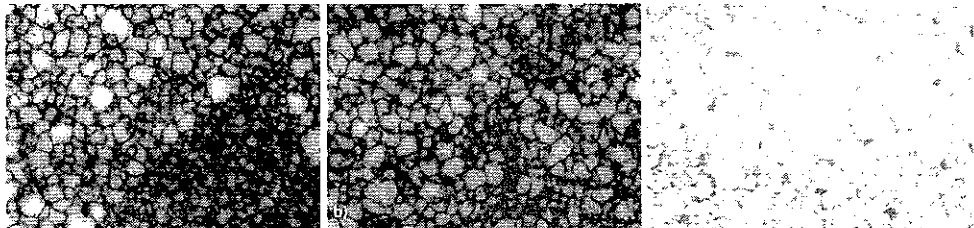
**Figure 5** Fuel-cladding bond and edge microstructure of fuel in etched condition at approx. 1073 mm from the bottom end. X1000 etched



**Figure 6** Transverse micrograph of section taken from 1632 mm from bottom end of rod E. a) X6 as polished. b) X6 etched (note central core)



**Figure 7.** Low power view of fuel microstructure: a) porous outer rim (no visible grain boundaries). b)  $\approx$  mid-radius. (X168 etched) (rod E)



**Figure 8** Microstructure at 1632 mm from bottom end. a)  $\approx$  radius note region of very small grain; etched). b) just inside of mid-radius position. c) fuel centre well developed intragranular pores. (X1000 etched) (rod E)

Radial composition profiles for certain elements (Pu, Nd, Xe, Cs, Mo and I) are shown graphically in Figure 9 for rod E at 1091 mm sample position. Nd and Zr in the case of rod E show similar, burnup related profiles. The Zr profile for the 1632 mm sample position does, however, exhibit a slightly decreasing gradient away from the rim, compared to the others; the Zr level measured at the rim for the sample at 1632 mm shows an "unexpectedly" high level of 26.12 %, which is due to the area being sampled having encompassed part of the fuel-cladding bonding layer.

Xe concentrations in both samples show a characteristic drop towards the rim associated with enhanced porosity in this region. The Cs concentration profile for the 1632 mm sample position shows the same constant level and rim enhancement as for Pu, Nd, and Zr. This suggests that very little redistribution has occurred. The corresponding profile for the 1091 mm position shows the same general trend, but with several enhanced concentration "spikes" across the radius. This would suggest some degree of Cs redistribution. Mo and Ru profiles for both samples again showed the same fairly constant level across most of the radius with enhanced concentration close to the rim. This suggests that these elements are present primarily in the fuel matrix, rather than as discrete metallic particles.

### 2.1.2.3. Fission gas release

Two strong trends are apparent on the fission gas data: power history and burnup effects. The low-power rods from groups C and D (standard enrichment) have consistently low gas releases (< 2 %) up to burnups of  $\approx 40$  MWd/kg U. By contrast, the high-power rods (high enrichment) show significantly higher gas releases ( $\approx 7$ -13 %) at comparable burnups of 35 - 40 MWd/kg U. Significantly, the higher power fuel microstructure show higher levels of grain growth and gas bubble formation along grain boundaries than the low-power fuel that operated at lower temperatures.

The burnup factor is shown most strongly by the general trend apparent in the Group A and B data for high-power rods. The gas release increases from 1 - 5 % at low burnups of  $\approx 30$  MWd/kg U to 13 - 27 % at high burnups of  $\approx 55$  MWd/kg U. Fission gas sensitivity to burnup is evident only in the high-power rods. The fuel rod design parameters of pre-pressurised with He and differences in initial fuel density have little apparent effect on gas release (Figures 10 & 11).

Table 3 lists the results of the mass spectrometric gas analysis, along with the voidage, internal pressure and gas volume for rods D (burnup 49.6 MWd/kg U), (burnup 57.2 MWd/kg U) and F (burnup 58.2 MWd/kg U) after cycle 5.

For rod D, the compositional analysis appears higher than expected for a He filled, pressurised rod, with the voidage and gas volumes being close to anticipated. For rod E, however, which is reported to be an air-filled, non-pressurised rod, the results are not as expected. The low volume, high He content and higher than expected air content, are suggestive of a leak somewhere in the system and/or contamination of extracted gas sample.

Other postulation is that there may have been a small leak present in the rod end- cap which allowed ingress of the during pressure testing. However, if such a leak should existed, the levels of fission gas observed for rod E would not have been expected.

**Table 3** Fission gas analysis results following puncture of rod D, rod E and rod F

Analyte	Composition (% V/V)		
	Rod D	Rod E	Rod F
Ar	0.011 $\pm$ 0.001	0.34 $\pm$ 0.002	0.003 $\pm$ 0.001
CH <sub>4</sub>	No detected	No detected	No detected
CO <sub>2</sub>	0.017 $\pm$ 0.003	0.019 $\pm$ 0.003	0.003 $\pm$ 0.002
H <sub>2</sub>	0.001 $\pm$ 0.002	0.002 $\pm$ 0.002	No detected
He	91.27 $\pm$ 0.16	60.34 $\pm$ 0.16	93.48 $\pm$ 0.16
<sup>84</sup> Kr / <sup>85</sup> Kr	0.177 $\pm$ 0.001	0.171 $\pm$ 0.001	0.165 $\pm$ 0.001
<sup>84</sup> Kr / <sup>86</sup> Kr	0.640 $\pm$ 0.001	0.673 $\pm$ 0.001	0.681 $\pm$ 0.001
<sup>80</sup> Kr / <sup>86</sup> Kr	0.027 $\pm$ 0.001	0.027 $\pm$ 0.001	0.026 $\pm$ 0.001
N <sub>2</sub> + CO	0.90 $\pm$ 0.01	29.94 $\pm$ 0.04	0.25 $\pm$ 0.01
N <sub>2</sub> O	Not detected	No detected	No detected
NO	Not detected	No detected	No detected

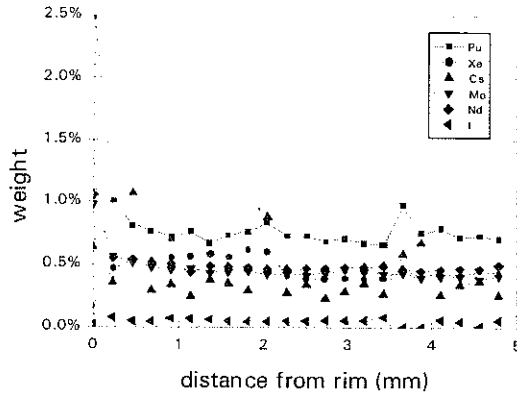


Figure 9 Concentration profiles for 1091 mm sample position (rod E)

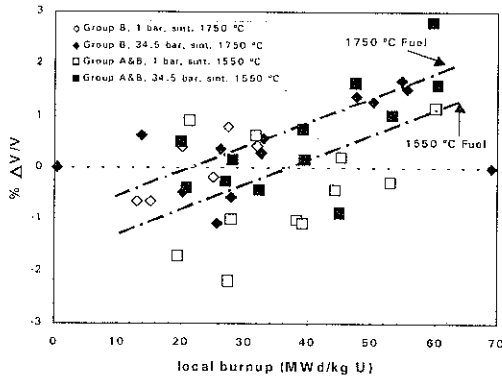


Figure 10 Fuel swelling as a function of burnup

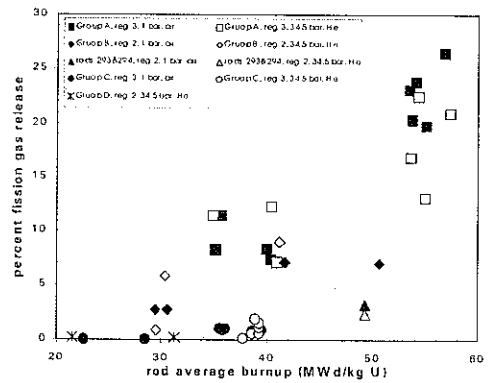


Figure 11 Fission gas release vs. fuel burnup

Analyte	Composition (% V/V)		
	Rod D	Rod E	Rod F
O <sub>2</sub>	0.20 ± 0.01	8.39 ± 0.01	0.06 ± 0.01
Total Kr	0.85 ± 0.02	0.09 ± 0.01	0.56 ± 0.01
Total Xe	6.74 ± 0.13	0.87 ± 0.01	5.64 ± 0.11
Xe / Kr	7.93	9.67	10.07
<sup>131</sup> Xe / <sup>134</sup> Xe	0.226 ± 0.001	0.214 ± 0.001	0.202 ± 0.001
<sup>132</sup> Xe / <sup>134</sup> Xe	0.772 ± 0.002	0.825 ± 0.002	0.839 ± 0.002
<sup>134</sup> Xe	1.93 ± 0.03	0.24 ± 0.01	1.58 ± 0.02
<sup>136</sup> Xe / <sup>134</sup> Xe	1.483 ± 0.003	1.497 ± 0.003	1.52 ± 0.03
<sup>130</sup> Xe	0.023 ± 0.01	0.002 ± 0.01	0.017 ± 0.01
Rod voidage (ml at NTP)	24.13	9.08	12.76
Rod internal pressure (bar abs.)	45.62	9.45	45.77
Gas volume (ml at NTP)	1101	86	583.0

Following this result, the end-cap region was re-examined visually, and no defect was observed. The integrity of the end cap region was also tested by pressurisation, and found to be sound. It may therefore be concluded that the rod was sound.

The measured rod voidage was also lower than expected. This was re-measured, however, and the result confirmed.

#### 2.1.2.4. Clad mechanical testing

Clad samples from three-cycles were mechanically tested. Closed burst tests were performed at 315.5 °C on 203 mm clad sections with fuel left intact. The samples were pressurised with hydraulic fluid at a rate of ~ 138 bar per minute. Tensile tests were performed at 315.5 °C on 152.4 mm long tube sections with most of the fuel mechanically removed. The relative strain rate up to yield point was  $8.33 \cdot 10^{-5} \text{ s}^{-1}$  and  $8.33 \cdot 10^{-4} \text{ s}^{-1}$  for the remainder of the test.

Three tube burst test samples and two tensile test samples, 200 mm in length, were cut from rod E. All the samples were completely defueled before testing. The samples are summarised in Table 4. Time/pressure values for burst tests were taken from the test data. The burst test results are presented in Table 5. The post-test appearance of the burst test samples is shown in Figure 12.

**Table 4** Summary of dimensional measurements on rod E cladding samples

Sample Identity	Position	Test type	Outer diameter (mm)		Inner diameter (mm)	
1	0857-1057	Tensile			9.32	9.25
2	1405-1605	Tensile			9.32	9.25
3	0657-0857	Burst	10.74	10.74	9.32	9.32
4	1110-1310	Burst	10.77	10.77	9.30	9.30
5	1405-1605	Burst	10.77	10.77	9.37	9.35
			Mean = 10.76		Mean = 9.31	

**Table 5** Tube burst test result from rod E

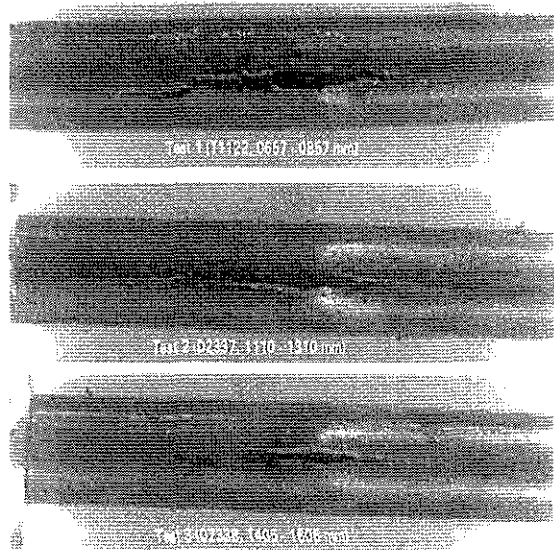
Sample Identity	Position (mm)	Burst Pressure (MPa)
T1122 (48)	0657-0857	137.4
D2337 (49)	1110-1310	136.3
D2338 (50)	1405-1605	115.4

Table 6 shows the mechanical properties of Zry clad in function of number of cycles.

**Table 6** Evolution of the clad mechanical properties as a function of number of cycles.

	preirradiated	Three cycles	Five cycles
Total elongation (%)	14.0 – 15.0	3.1 – 6.0	5.40 – 7.40
Ultimate tensile strength (MPa)	720.50 – 742.56	698.44 – 774.28	819.50 – 856.10
0.2 % proof load stress (MPa)	524.69 – 539.86	563.30 – 643.97	687.80 – 724.40

The overall mechanical test results indicate that rods cladding retained adequate strength (yield stress) and ductility (uniform elongation) through three cycles of exposure to fast neutron fluency in excess of  $5 \cdot 10^{21} \text{ n/cm}^2$ .



**Figure 12** Appearance of tube burst pieces

### **3. Conclusions**

After more than twenty years of dry storage the PWR rods not present a signify anomalies, comparing it with the tests carried out before its storage.

### **4. Acknowledgements**

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