THE POST-IRRADIATION EXAMINATION OF CANDU TYPE FUEL IRRADIATED IN THE INSTITUTE FOR NUCLEAR RESEARCH TRIGA REACTOR

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ABSTRACT

This post-irradiation examination work has been done under the Research Contract No. 7756/RB, concluded between the International Atomic Energy Agency and the Institute for Nuclear Research. The paper contains a general description of the INR post-irradiation facility and methods and the relevant post-irradiation examination results obtained from an irradiated experimental CANDU type fuel element designed, manufactured and tested by INR in a power ramp test in the 100 kW Pressurised Water Irradiation Loop of the TRIGA 14 MW(th) Reactor. The irradiation experiment consisted in testing an assembly of six fuel elements, designed to reach a burnup of approx. 200 MWh/kgU, with typical CANDU linear power and ramp rate.

1. INTRODUCTION

The Institute of Nuclear Research in Pitesti has a set of nuclear facilities that allow various investigations on CANDU type nuclear fuel and materials:

- TRIGA 14 MW(th) Materials Testing Reactor;
- Irradiation loops and capsules;
- Post-Irradiation Examination Laboratory.

The main irradiation tests carried out on experimental fuel elements are power ramps, power cycling, fission gas pressure evolution and fission gas release rate evolution.

The Post-Irradiation Examination Laboratory (PIEL) is an alpha-gamma hot cell facility, commissioned and licensed for nuclear operation in 1984. This facility has the capability to carry out post-irradiation examination (PIE) on irradiated reactor fuel and structural materials in support to the INR nuclear fuel research and development programme.

The objective of the paper is to present the relevant results obtained by post-irradiation examination of a CANDU type experimental fuel element, tested in a small assembly of six rods in a power ramp test. The test was carried out in the TRIGA reactor, in one of the existing irradiation device.

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2. DESIGN AND MANUFACTURING DATA OF THE EXPERIMENTAL FUEL ELEMENT

The experiment was designed by the INR group responsible for the development of computer codes for the analysis of fuel performance in different operation conditions. The main objective of the experiment was to verify the behaviour of experimental CANDU type fuel elements during power ramps at burnups beyond 160 MWh/kgU. In this scope the six experimental fuel elements were designed, with a reduced length of 266 mm and with 5wt% enriched UO_2 pellets, in order to achieve an uniform axial distribution of the neutronic flux and respectively a significant linear power required by this type of test. Flux suppressers were not provided at the ends of the fuel column. The specified irradiation parameters were:

- Element burnup [MWh/kgU]: 200
- Element linear power, low power phase [kW/m]: 35
- Element linear power, after ramp [kW/m]: 58
- Ramp rate [kW/m.s]: 0.025

The experimental fuel elements were manufactured at INR, by a specialised group, and the design specifications were fulfilled.

3. IRRADIATION HISTORY

The irradiation experiment has been conducted in the 100 kW Pressurised Water Irradiation Loop, installed in the core of the TRIGA reactor. The power ramp has been performed at a burnup of 185 MWh/kgU. The total irradiation time has been of 6048 hours. The specified irradiation parameters were also respected, as this fact was demonstrated by the inferred values from the reactor and irradiation device instrumentation records. The evaluated burnup at the end of the irradiation was 206 MWh/kgU, with an accuracy of $\pm$ 10%.

4. POST-IRRADIATION EXAMINATION

4.1. Methods and techniques for post-irradiation examination.

The heart of the facility consists of a block of two concrete shielded hot cells (37 PBq), equipped with nine working stations. Adjacent to these cells is a smaller block of two steel shielded cells (37 TBq), equipped with two working stations, used for metallographic examination and for chemical operations. Fast transfer of samples between the concrete cells and the steel shielded cells is done by a pneumatic rabbit.

The post-irradiation examination methods and their associated techniques were developed by the PIEL staff, based on a standard reference recommended practice for post-irradiation examination of water reactor fuel elements [1]. The available PIE methods are:

- Visual inspection and photography
- Dimensional control (diameter, bow, length)
- Eddy current control for clad integrity
- Axial and radial gamma scanning, tomographic 3-D examination
- Gas pressure and volume and void volume determination
- Metallography, ceramography and quantitative microscopy

- Radiochemistry and burnup determination
- Mechanical testing at room temperature and at 300 °C

In connection with these methods, the following typical techniques are used: dismantling/reassembling, visual inspection by a monocular periscopes, profilometry, gamma spectrometry, eddy current testing, mechanical puncturing and fission gas measurement, precision low speed diamond disc cutting, metallographic preparation, microscopy , microhardness testing, fuel sample dissolution, alpha and gamma spectrometry, mass spectrometry, tensile and burst testing of cladding, interim storage of conditioned fuel fragments in storage pits, treatment and conditioning of solid wastes resulting from the process.

The non-destructive examination process is performed by the use of three computer controllable universal examination machines, which have in principle the same basic design. During the examination process the fuel elements have a vertical atitude and the same axial and azimuthal origin. Before starting a control process, manual verification of the calibration is performed, using for this purpose adequate calibration gauges.

The macrographic examinations and detailed microscopic examinations are done using a LEITZ 5 MM RT microscope and his associated image analyser. The radiochemical process occurs in the radiochemistry sublaboratory, equipped with a high resolution gamma spectrometer, an alpha spectrometer and a FINNIGAN MAT 261 mass spectrometer.

4.1. Non-destructive examination

Visual control and photography. (see Fig.1) The control operation was done along the fuel element axis and the outer surface was photographed at three azimuthal positions (0°, 120° and 240°). The control has no revealed particular items to be considered later (i.e. defects, deposits, corrosion effects and excessive dimensional changes). The obtained images, show clearly that corrosion rate in the heat affected zone of the brazed appendices, containing preponderantly the beta structural phase of zirconium, was higher than on the rest of the sheath.

Dimensional control. The control operation has been performed, both before irradiation and after irradiation, in order to have the possibility to calculate the ratio of dimensional changes. The fuel element profile along the axis has been measured at three azimuthal positions (0°, 120° and 240°) and at regular intervals of 0.001 m. The LVDT transducers have been calibrated using a standard diameter gauge of 13.08 $\pm$ 0.04 mm. The dimensional control revealed the following results:

- Diametral deformation at pellet interface, mean value [%]: 0.83
- Diametral deformation at pellet midplane, mean value [%]: 0.43
- Overall diametral deformation, mean value [%]: 0.43
- Bow, mean value [mm]: 0.105
- Axial elongation, mean value [%]: 0.03

Eddy current control for clad integrity. This control operation has been done in order to detect different imperfections and defects in the cladding material and has confirmed the integrity of the fuel element. In order to obtain a maximum sensitivity for detection and to adjust the reference phase for the current the calibration has been done using an artificial

defect gauge, containing artificially produced defects such as internal and external axial slits (1250 x 10 x 100 microns) and penetrating holes (320, 230, 110 microns in diameter).

Axial gamma scanning (see Fig.1). This operation was used for axial discrete acquisition of specific gamma spectra of Cs-137 (661.643 keg), Zr-95 (724.23 keV) and La-140 (1596.17 keV), shown as integral activity axial profile. In this scope a collimator slit opening of 0.0005 m and a discrete displacement 0.0005 mm were used. The calibration for energy is performed using standard Co-60 and Cs-137 sources, while the calibration for efficiency uses Sb-124 and Cs-137 sources. The increase of activity at the ends of the fuel column is caused by the end peaking of the neutron flux. Based on the fact that no axial migration of Cs-137 was observed and that radionuclide is a long life and has practically the same fission yield both for U-235 and Pu-239, burnup calculation has been done by using the measured activity.

The "N" number of existent Cs-137 atoms has been calculated by the use of the relationship:

$$N = \frac{\sum_i A_i}{\varepsilon \cdot K_{auto} \cdot s \cdot t \cdot \lambda} \qquad (1)$$

where:

- A_i - area of the peak recorded from the fuel rod at "I" axial elevation;
- ε - efficiency of detection;
- K_{auto} - gamma self-absorption coefficient;
- s - gamma-ray abundance;
- t [s] - time of acquisition;
- λ [s^{-1}] - decay constant.

For the calculation of the burnup "BU" the following relationship has been used:

$$BU[MWh / kgU] = \frac{N \cdot E_f}{\eta_{Cs} \cdot m_U} \cdot 4.45 \cdot 10^{-20} \qquad (2)$$

where:

- η_{Cs} - fission yield of Cs137;
- E_f [MeV] - fission energy;
- m_U [g] - mass of metallic uranium in the fuel rod;
- 4.45×10^{-20} - transformation coefficient (1 MeV/gU to 1 MWh/kgU).

The calculated value for burnup is 188.4 MWh / kgU (192 MeV/fission), with an estimated accuracy of 10 %.

Tomographic 3-D examination (see Fig.2). In order to obtain the spatial distribution of activity of Cs-137 and La-140, in a cross section of the fuel element, the measured radial profiles were used in conjunction with a method of tomographic reconstruction based on a maximum entropy algorithm [2]. The radial gamma spectra acquisitions were done at five angular positions (0°, 72°,144°, 216°, 288°) and at a regular movement of 0.00025 mm of the element in front of the collimator, having a slit of 0.005 m in height and of 0.00025 m in the aperture. The distribution of La-140 shows some agglomeration of this radionuclide in the centre of the fuel pellet.

4.2. Destructive examination

Fuel rod puncture and fission gas measurements. Fuel rod puncturing and fission gas pressure and inner free volume measurements are carried out using an appropriate device. The technique is based on the puncture of the fuel rod in vacuum and a subsequent pressure measurement, followed by introduction in the system of a known volume of inert gas and pressure measurement. The obtained results are:

- Fuel rod inner pressure [kPa]: 488, accuracy 2%
- Gas volume [cm^3]: 4.78, accuracy 5%
- Fuel rod inner free volume [cm^3]: 1.25, accuracy 5%

Metallography and ceramography. Metallographic and ceramographic examination was based on the cutting plan shown in Fig.1. Four metallographic samples M1, M2, M3, M4 were prepared and examined. The sample cutting and preparation follows a typical procedure, based on resin impregnation of the fuel element, slow speed diamond disk precise cutting, sample embeddment in cold curing resin, metallographic preparation, ultrasonic bath cleaning and surface quality inspection by a periscope.

General view of the sample (see Fig.1). In this scope macrography was performed at 3x magnification and the corresponding images are shown in Fig.1. Due to the end flux picking, the M4 sample (bottom end of the fuel element) shows serious restructuring with central hole and with the central part of fuel pellet displaced toward the end plug, while the other three samples show a typical irradiated fuel macrostructure.

Dimensional measurements. This control operation has been done both in order to compare local results with non-destructive dimensional control results and to reveal some other characteristics, like gap dimensions and distribution or metallographic parameters like grain size, structural zone radius and plastic deformation radius. For the first case the measurements are performed using the discrete displacement of microscope stage with an accuracy of $\pm$ 10 micrometers . For metallographic parameter determinations the measurements are performed by the use of the micrometric scale wit 0.5 micrometer division, incorporated in the special ocular of the microscope. The dimensional measurements on M2 sample revealed an excellent correspondence between the results obtained by non-destructive dimensional control and outer diameter measurements.

Cladding surface condition (see Fig.3a). The outer surface of the cladding was found uniformly covered with an adherent layer of ZrO_2 with a thickness of 2-3 micrometers . The inner surface of the cladding was found generally clean, free of scratches or fuel indentations. Exceptions were found, near the interface between pellets and sheath ends, having a form of short oxide layer (lens oxide layer) with a length smaller than 200 micrometers. These oxide formations showed a tendency to connect themselves and to form a continuous layer, having a thickness of 5-7 micrometers

Cladding structure (see Fig.3b). The hydriding condition of the cladding is shown on M2 sample A content of hydrogen of about 80 ppm was estimated by means of hydruration charts.

Condition of brazed and welded zones (see Fig.3c). The condition of brazed zones (plugs, bearing pads, spacers) was also extensively examined. A brazing defect was found on sample M2 at the interface between the bearing pad and the cladding. The end plug weld was found as satisfactory. The Vickers hardness values obtained were about 260-280 units in the HAZ and about 220-240 units at great distance from this zone.

Uranium Dioxide Microstructure (see Fig. 4a and 4b). The UO_2 microstructure was revealed by chemical attack with a mixture of H_2SO_4 and H_2O_2. Three of the metallographic samples (M1, M2, M3) contain only two zone structure, equiaxed grown grains and as-sintered grains. The M4 sample had shown a three zone structure with columnar, equiaxed and as-sintered grains zones. The measurement of the radii of the two zones on M2 sample indicated the following values:

- Equiaxed grown grains radius [mm]: 3.1 ± 0.02
- Pellet radius [mm]: 6.13 ± 0.02

The evaluation of the grain size has been done by the use of the interception method [3]. The obtained equivalent ASTM numbers for grain size were:

- Grains, as sintered [micrometers]: 7.9 ± 0.1
- Grains, equiaxed [micrometers]: 20 ± 0.2

4.3. Fuel burnup determination by mass spectrometry.

The absolute burnup value attained by the experimental fuel element has been determined by the use of the method of atom percent fission in uranium fuel [4]. The mass spectrometric determination of the concentrations and isotopic abundance have been performed on irradiated and nonirradiated processed fuel sample solutions having a concentration of approximately 1 gU / L. The obtained burnup value is 194.3 $\pm$ 5.9 MWh / kg U, with an accuracy $\pm$ 3.0 %, and was obtained by the use of rigorous procedures for data correction and burnup calculation.

5.0. SUMMARY

The paper summarises the general procedure and results of a typical PIE work carried out on an experimental CANDU type fuel rod, designed, fabricated, irradiated and investigated for performance at INR Pitesti in the existing nuclear facilities.

The examined fuel element was tested in a power ramp test, designed for a burnup of 200 MWh/kgU and for power levels of 35 kW/m in low power phase of operation and of 58 kW/m after the ramp, with a ramp rate of 0.025 kW/m.s. The fuel element has attained practically a burnup of 194.3 ($\pm$3%) MWh/kgU, value determined by mass spectrometric method, and revealed integrity, stability and a behaviour as it was predicted for such operating conditions by the group responsible for experiment design and calibration of computer codes for fuel performance analysis.

REFERENCES

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(2) A. ALEXA, T. CRACIUNESCU, G. MARINESCU, R. DOBRIN, "The tomographic maximum entropy method in the 3-D analysis of nuclear fuel elements", Journal of Nuclear Materials 218 (1995) 139-142

(3) H. ABRAMS, "Practical Applications of Quantitative Metallography", Stereology and Quantitative Metallography, ASTM STP 504, American Society for Testing and Materials, 1972, pp.138-182

(4) AMERICAN NATIONAL STANDARD INSTITUTE, ASTM E 244 - "Standard Method of Test for Atom Percent Fission in Uranium and Plutonium Fuel (Mass Spectrometric Method)"

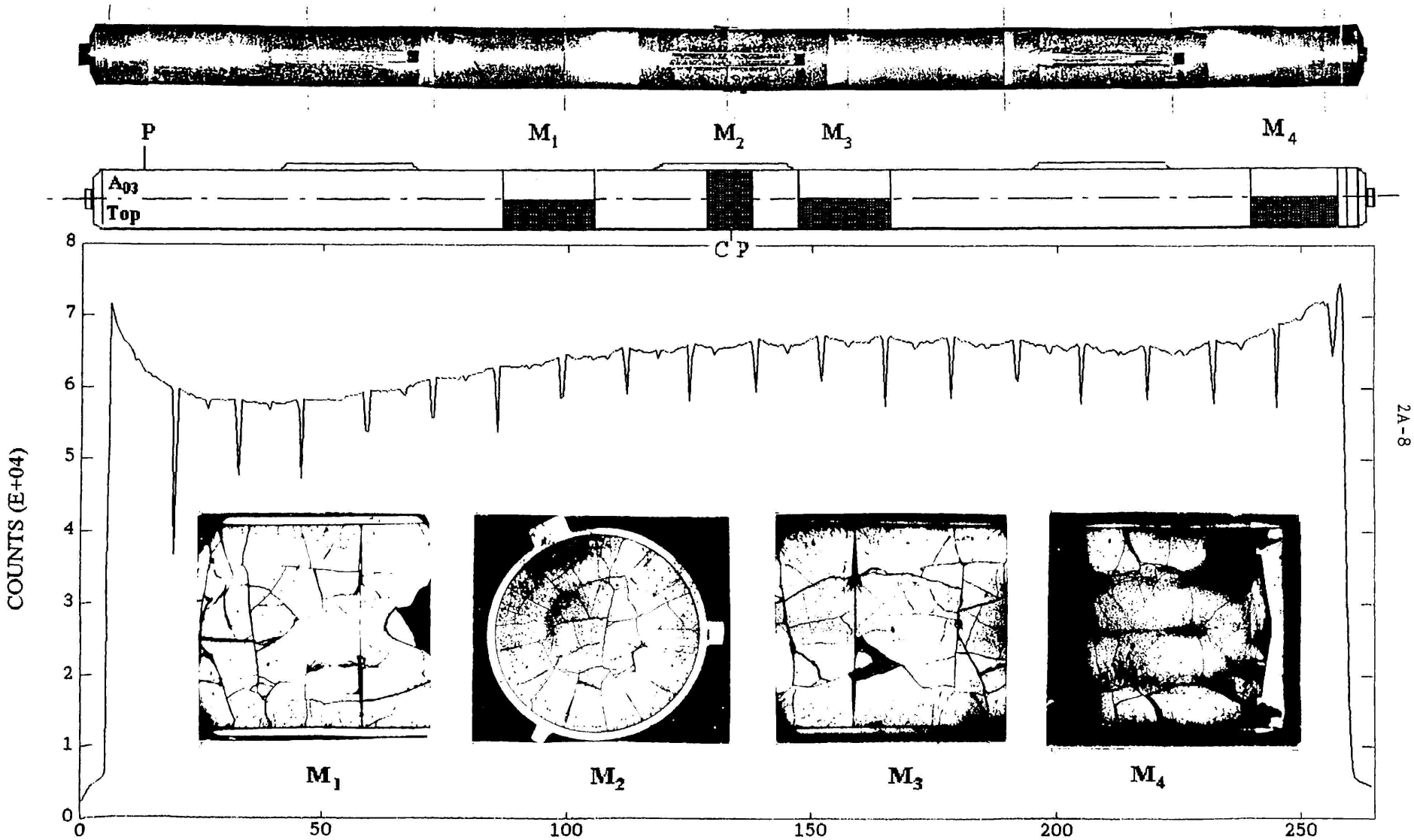


Fig. 1 Axial gamma scan from A_{03} fuel rod. Integral profile.

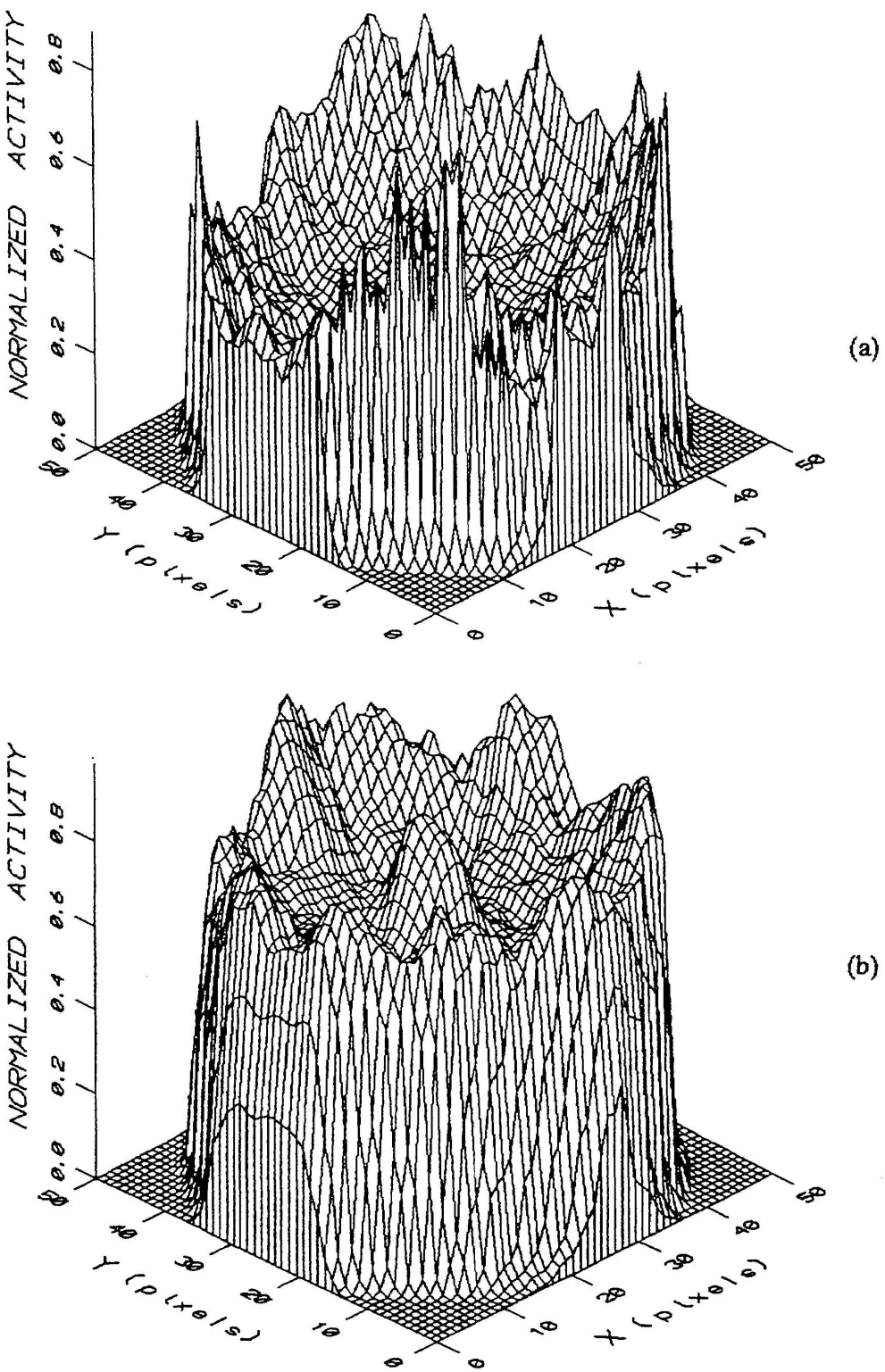


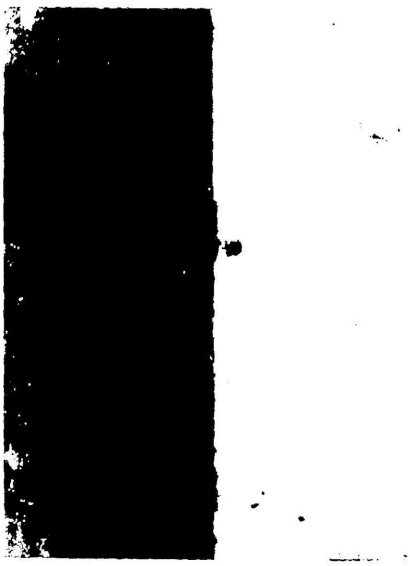
Fig. 2 Reconstruction of the ^{137}Cs and ^{140}La distribution in the cross section of the fuel rod

a - ^{137}Cs

b - ^{140}La

(a)

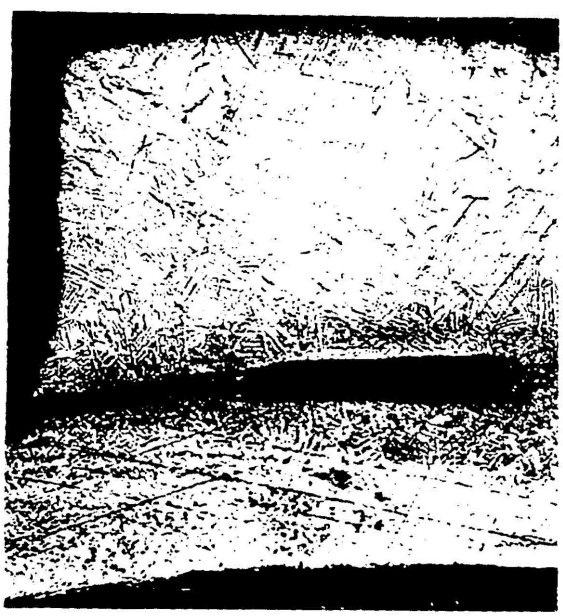
(b)



(a) Hydride plates (200 x)

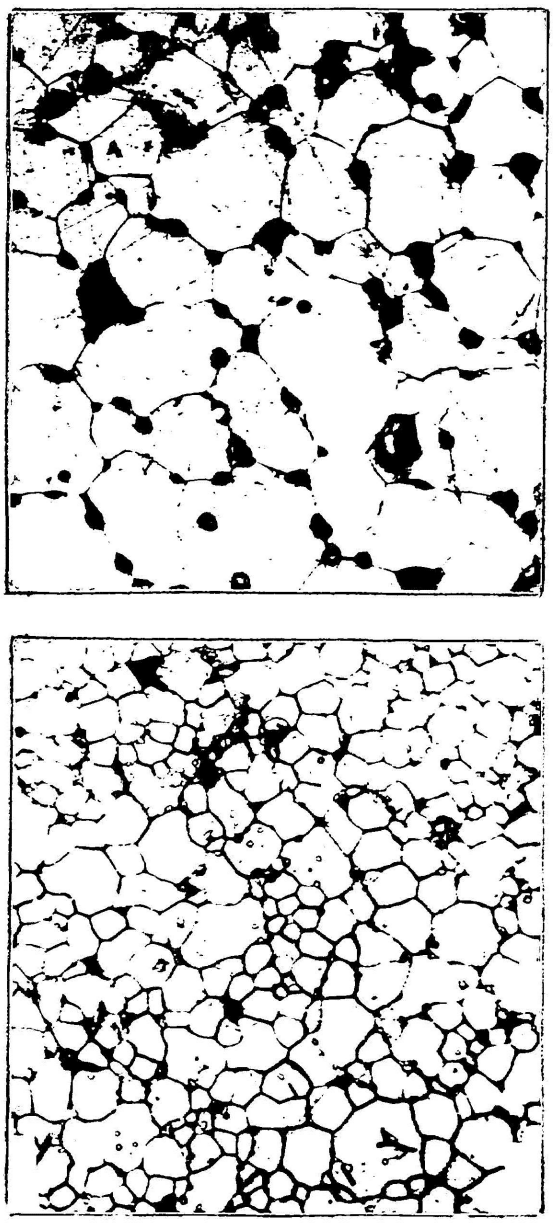
(b) Inner oxide layer (400 x)

(c) Pore in pad-clad brazed zone (65 x)



(c)

Fig. 3 Clad microscopic examination



a

b

Fig. 4 Fuel microstructure (500x)
a - Equiaxed grown grains
b - As sintered grains