Available Post-Irradiation Examination Techniques at Romanian Institute for Nuclear Research

Marcel PARVAN[1], Antonius SORESCU, Marin MINCU, Octavian UTA, Relu DOBRIN
Institute for Nuclear Research, Post-Irradiation Examination Laboratory
110000, Piteşti, P.O.Box 78, Romania

Abstract

The Romanian Institute for Nuclear Research (INR) has a set of nuclear facilities consisting of TRIGA 14 MW(th) materials testing reactor and LEPI (Romanian acronym for post-irradiation examination laboratory) which enable to investigate the behaviour of the nuclear fuel and materials under various irradiation conditions.
The available techniques of post-irradiation examination (PIE) and purposes of PIE for CANDU reactor fuel are as follows: - Visual inspection and photography by periscope: to examine the surface condition such as deposits, corrosion etc; - Eddy current testing: to verify the cladding integrity; - Profilometry and length measurement performed both before and after irradiation: to measure the parameters which highlight the dimensional changes i.e. diameter, length, diametral and axial sheath deformation, circumferential sheath ridging height, bow and ovality; - Gamma scanning and Tomography: to determine the burnup, axial and radial fission products activity distribution and to check for flux peaking and loading homogeneity; - Puncture test: to measure the presure, volume and composition of fission gas and the inner free volume; - Optical microscopy: to highlight the structural changes and hydriding, to examine the condition of the fuel-sheath interface and to measure the oxide thickness and Vickers microhardness; - Mass spectrometry: to measure the burnup; - Tensile testing: to check the mechanical properties. So far, non-destructive and destructive post-irradiation examinations have been performed on a significant number of CANDU fuel rods (about 100) manufactured by INR and irradiated to different power histories in the INR 14 MW(th) TRIGA reactor. These examinations have been performed as part of the Romanian research programme for the manufacturing, development and safety of the CANDU fuel. The paper describes the PIE techniques and some results.

Keywords: *hot laboratory, hot cell, post-irradiation examination, eddy current, profilometry, tomography, fission gas analysis, metallographic examination, microhardness, burnup*

Introduction

The Romanian Institute for Nuclear Research by its Post-Irradiation Examination Laboratory (LEPI) performs PIE /1/ on nuclear fuel and materials in the Hot Cells since 1984. The LEPI is a hot laboratory brought into operation in December 1983. It includes two heavy concrete hot cells, three steel hot cells and one lead hot cell. The LEPI is located adjacent to the TRIGA materials testing reactor and a water canal used to transfer the irradiation experiments from the TRIGA reactor pool to the hot cell interconnects both.

The PIE objective is to provide reliable results on the cladding integrity, surface condition, dimensional changes, fission products distribution, fission gas release, hydriding, structural changes, mechanical properties and burn-up in order to monitor the performance of the CANDU-6 fuel in operation.

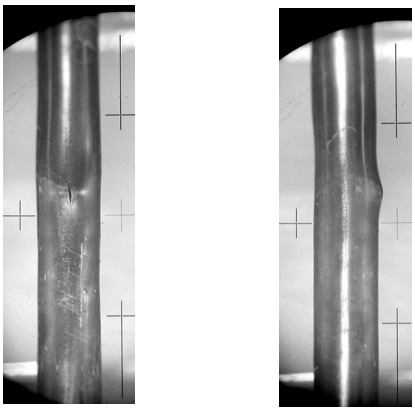
Non-destructive PIE techniques

The non-destructive examination (NDE) techniques usually applied to the irradiated fuel rods include visual inspection and photography, dimensional measurements, gamma scanning, tomography and eddy current testing. The main equipments for NDE consist of three fuel rod-positioning machine: one for visual inspection, one for profilometry and eddy current testing and one for gamma scanning.

Visual inspection and photography

A CLAVE model 291A periscope set in the hot cell shielding wall and an in cell fuel rod-positioning machine are used to examine the irradiated fuel rod. The periscope has 4 magnifications: x2.1; x4.2; x5.9; x11.6 and it is equipped with a HASSELBLAD photographic camera (size 6x6) for taking of photographs. Also, an OLYMPUSdigital camera is used to take photos by the periscope eyepiece.
The macroscopic characteristics of the cladding such as deposits, corrosion, cracks, swelling are examined by periscope and photos are taken (Fig. 1).

[1] Tel: (+40) 248 21 34 00, mparvan@scn.ro



Rotation 0° Rotation 90°

Fig. 1 CANDU fuel rod cracked during a RIA test in TRIGA pulse reactor

Profilometry

The profilometry is a step-by-step measurement of diameter along the fuel rod at regular intervals (usually 1 mm) using a remotely operable profilometer. The data are used to determine the parameters that highlight the dimensional changes of the fuel rod during irradiation: diameter, length, diametral and axial sheath deformation, circumferential sheath ridging height, bow and ovality.
The profilometer (Fig. 2) consists of a vertical fuel rod-positioning machine equipped with SLO-SYN step-by-step motors, a measuring device using two opposed SCHLUMBERGER inductive transducers as sensors (as shown in Figure 3) and a control desk equipped with digitally pulse counters to display the fuel rod position. It is piloted by computer.

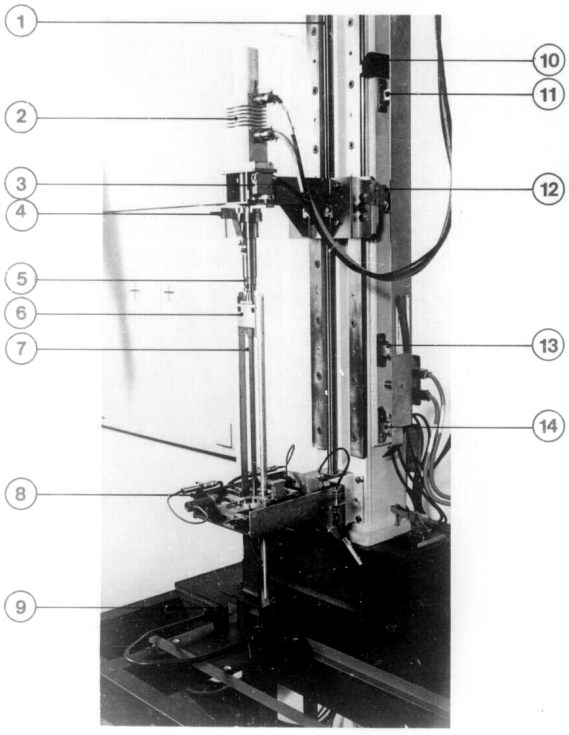


Fig. 2 Profilometer for CANDU fuel rod diameter measurement

1 – Screw for "Z" vertical movement
2 – Step-by-step motor for "R" rotational movement
3 – "R origin" microswitch
4 – Gripping device lever
5 – Gripping device
6 – Fuel rod holder
7 – Fuel rod
8 – Diameter and bow measuring device
9 – Fuel rod distribution carriage
10 – "Z" mechanical limit stop
11 – "Z origin" microswitch
12 – Microswitch driving cam
13 – Microswitch for "End of measurement"
14 – Microswitch for "Position of fuel rod gripping"

The main characteristics of the fuel rod-positioning machine are:

- Vertical movement (Z): 1500 mm ±0.05 mm, positioning by step of 0.01 mm controled by synchro resolver;
- Horizontal movement (X and Y): 150 mm ±0.01 mm, positioning by step of 0.01 mm controled by synchro resolver;
- Rotation: continuous, positioning by step of 0.09° controled by synchro resolver.

The inductive transducers are differential transformers having a mobile core. In order to eliminate the error of measurement due to unlinearity of transducers, their linearization has been accomplished. The calibration of the transducers is carried out before to start the measurements using a standard diameter gauge of 13.08 mm (nominal diameter of the CANDU fuel rod). It is possible to measure the diameters of 9 to 17 mm within accuracy of ±5 μm.accuracy of ±5 μm.

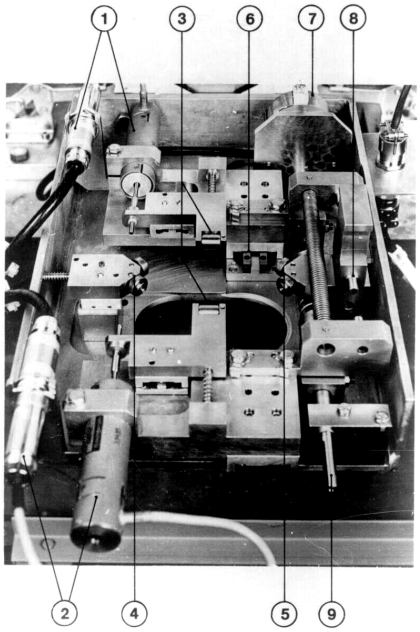
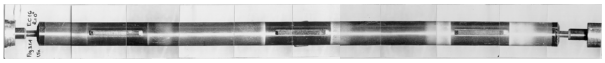


Fig. 3 Diameter measuring device of the irradiated CANDU fuel rod



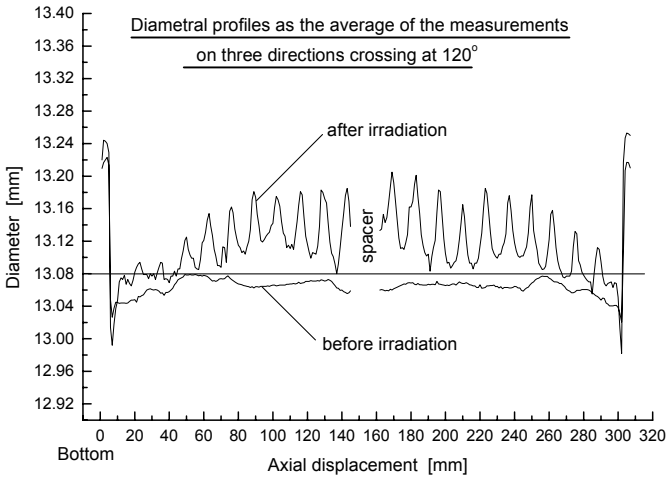


Fig. 4 Profilometry on a CANDU fuel rod irradiated in the INR TRIGA reactor in a power ramping test

1 – Transducer No.1 and its connector
2 – Transducer No.2 and its connector
3 – Transducer plungers
4 – Mobile guiding rollers
5 – Immobile guiding rollers
6 – Ball sliding carriage
7 – Motoreductor unit for transducer advancing/recoiling
8 – Gauge plug to limit transducer advancing
9 – Screw for carriage advancing/recoiling

The main technical characteristics of the transducers are:

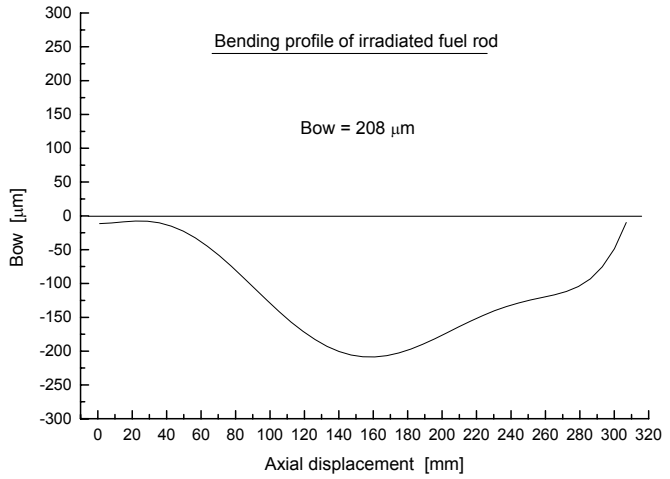
- Measurement range: ±2.5 mm
- Unlinearity error: ±0.5% of measuring range
- Maximum working temperature: 773 K (500°C)
- Sensibility variation depending on temperature: 0.0025%/K
- Environment: radioactive ($4 \cdot 10^{13}$ neutroni/cm^2·s)

The fuel rod is characterized dimensionally both before and after irradiation using the same equipments and procedures. The fuel rod diameter is measured on the three axial directions crossing at 120°. Figure 4 shows the diametral profile of a CANDU fuel rod before and after irradiation as the average of the three scans. The parameters which highlight the dimensional changes of this fuel rod are as follow:

- Average diameter before irradiation : 13.068 mm
- Average diameter after irradiation : 13.117 mm
- Number of measurements : 291
- Standard deviation : 3 μm
- Average diameter at top of ridges : 13.162 mm
- Average diameter at bottom of ridges : 13.086 mm
- Average height of the ridges : 38 μm
- Maximum diametral elongation : 145 μm (1,1 %)
- Length before irradiation : 307.04 mm
- Length after irradiation : 307.42 mm
- Axial elongation : 380 μm (0.1 %)

The bow of the fuel rod is determined by semi-difference of the transducers data (Fig. 4). For the accurate determination of the fuel rod ovality, circumferential diameter measurements are carried out. Figure 5 shows the circumferential diametral profile of a CANDU fuel rod irradiated in the TRIGA reactor. The dimensional parameters which highlight the ovalization of this fuel rod at 61 mm from bottom are as follow:

- Number of measurements : 20
- Rotational step : 9°
- Standard error : 3 μm
- Maximum diameter of ellipse : 13.363 mm
- Minimum diameter of ellipse : 13.310 mm
- Equivalent diameter of ellipse : 13.336 mm
- Average diameter before irradiation : 13.078 mm
- Circumferential elongation : 0,81 mm (2 %)



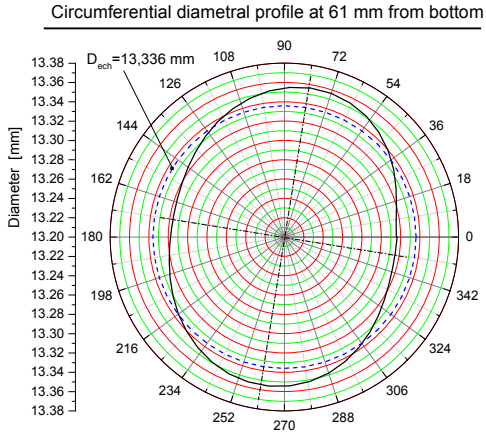


Fig. 5 Circumferential profilometry on a CANDU fuel rod irradiated in the INR TRIGA reactor in a overpower test

Gamma scanning and tomography

The gamma scanning technique is used to determine the axial and radial distribution of the fission products (FPs) activity and the migration of volatile FPs inside the fuel rod, to measure the fuel column length, to assess the gap between pellets and to calculate the burn-up. The gamma scanning system consists of a vertical fuel rod-positioning machine equipped with SLO-SYN step-by-step motors, a collimator set in the hot cell shielding wall, a PGT intrinsic Ge detector, a multichannel analyser using a CT 103 type 200 MHz ADC and 4096 channel memory and a control desk equipped with digitally pulse counters to display the fuel rod position. It is piloted by computer. The collimator includes three rectangular slits having the width of 50 mm and the aperture size of $0.1^{\pm 0.005}$ mm, $0.25^{\pm 0.005}$ mm and $0.5^{\pm 0.005}$ mm. The slits are made of tungsten alloy and their position can be horizontal or vertical. The absolute efficiency of detection at ^{137}Cs energy (661.66 keV) is obtained using a standard ^{137}Cs source (273 mCi ±4.5% to 18.08.1980). The fuel rod is axially scanned for gross gamma activity profile and isotopic gamma activity profiles (^{137}Cs, ^{134}Cs, ^{95}Zr-Nb, ^{103}Ru-Rh, ^{106}Ru-Rh, ^{140}Ba-La, ^{144}Ce-Pr). Figure 6 shows the axial gross gamma activity profile of a CANDU fuel rod irradiated in the TRIGA reactor. This profile enables to check for flux peaking and loading homogeneity.

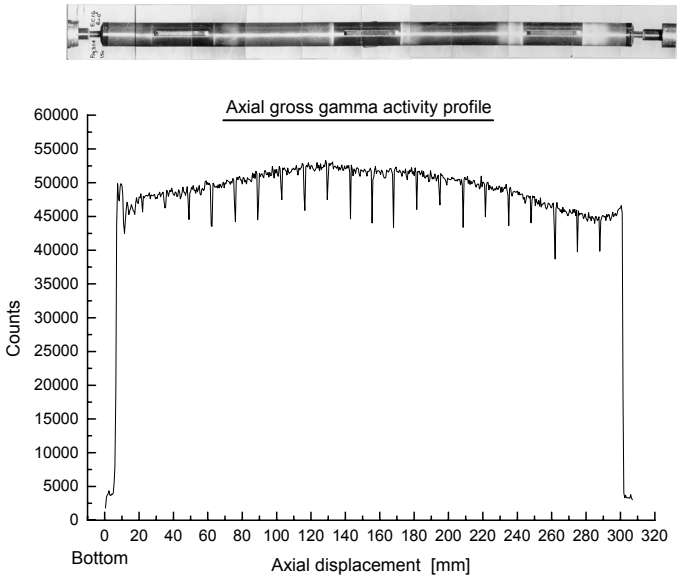
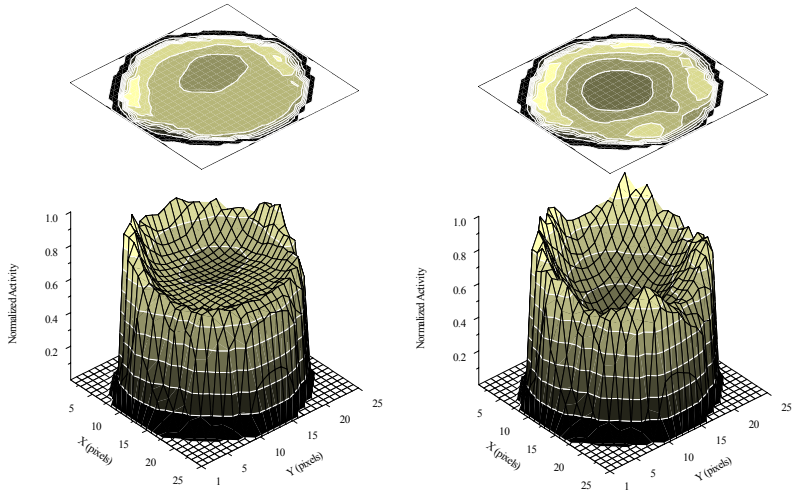


Fig. 6 Axial gamma scanning on a CANDU fuel rod irradiated in the INR TRIGA reactor in a power ramping test

The ^{137}Cs isotope is used as burnup monitor. For an accurate determination of the burnup, the gamma self-absorption coefficient must be calculated using the distribution of ^{137}Cs activity in the cross section of the fuel rod. This distribution is determinated by radial gamma scanning. The burnup of the fuel rod shown in Fig. 6 is 210.5 MWh/kgU (192 MeV/fission) with an accuracy of ±10 %. The fuel rod burnup determined by mass spectrometry is 216.15 MWh/kgU with an accuracy of ±3 %. These results are in agreement. The fuel rod enrichment is 5.75 wt% ^{235}U.

The irradiation conditions were:

- Linear power in pre-ramping: 43 kW/m;
- Linear power in ramping: 63 kW/m.



a) radial profile of ^{137}Cs isotope in the integrity zone of the sheath

b) radial profile of ^{137}Cs isotope in the defect zone of the sheath

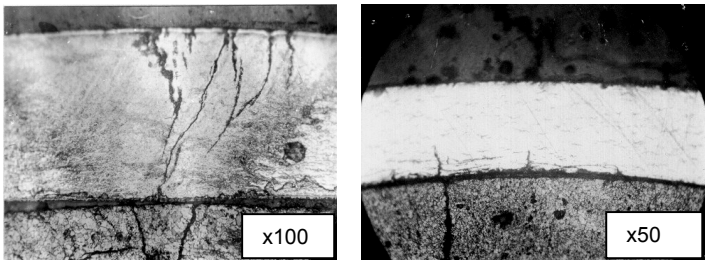
Fig. 7 Radial activity distribution of ^{137}Cs isotope in a CANDU fuel rod determined by tomographic method

A method of tomographic reconstruction based on a maximum entropy algorithm as it is described in /2/ has been developed. This method provides informations on the radial distribution of FPs activity in a cross section of the fuel rod (Fig. 7). The data acquisition is done while the fuel rod is moved transversally step-by-step at regular interval of 0.25 mm after every its rotation of 72^{o} in front of the collimator.
The tomographic image of the radial ^{137}Cs activity distribution shown in Fig. 7 b) indicates that this isotope migrated from middle to outside of the fuel rod and redistributed according to the temperature profile. The tomography has been used as complementary method for detection of the sheath defect. Fig. 7 is an example of this use /3/.

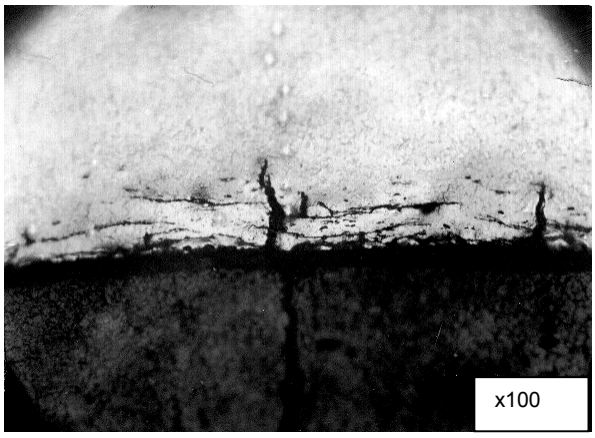
Eddy current testing

The eddy current technique provides informations on the integrity of irradiated fuel rod cladding. An INTERCONTRÔLE monochannel eddy current flaw detector operated in the test frequency ranging from 1 kHz to 1 MHz and an absolute probe coil mounted on the fuel rod-positioning machine are used. Defects of a few hundredths of a millimeter can be detected.

The empty Zircaloy-4 cladding tubes (diameter 13.08 mm, wall thickness 0.38 mm and length 500 mm) with defects artificially produced are used as the standard rods for calibration. The artificial defects are external and internal longitudinal and circumferential notches (width = 0.1 mm and depth of 25%, 50% and 100%) and holes (diameter of 0.1 mm, 0.2 mm, 0.3 mm and 0.4 mm). The calibration is accomplished to obtain the maximum sensitivity of detection and to adjust the phase of reference. The cladding defects detected by this technique have been confirmed by optical microscopy as shown in Fig. 8.



a) b)



c)

Fig. 8 Cracks in the irradiated CANDU fuel rod cladding detected by eddy current technique
a) Outer and penetrating cracks b) Inner cracks
c) Enlargement of the photo b)

Destructive PIE techniques

The destructive examination techniques usually applied to the irradiated fuel rods include fuel rod puncture test, optical microscopy, chemical analysis and burn-up determination, mechanical testing.

Fuel rod puncture test

This technique is used to measure the pressure and volume of fission gas inside the fuel rod and the fuel rod internal void volume. A SRS QMS 200 Gas Analyzer installed at the outside of the hot cell enables to analyse the fission gas composition.
The puncture tool installed in the hot cell is shown in Fig. 9. Prior to puncturing the fuel rod, the V1 volume of the various components in the system is measured. The technique used is to pressurize the V2 standard volume with helium at a measured pressure and to expand it into the V1 volume.

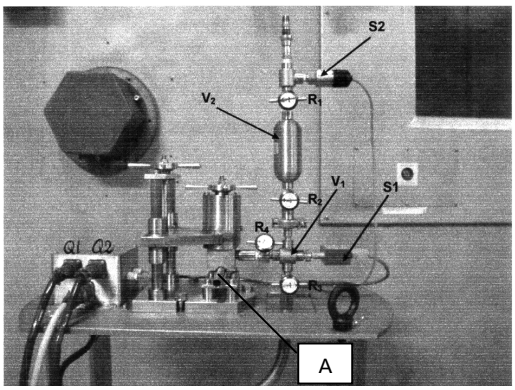


Fig. 9 Puncture tool

The equilibrium expansion pressure is measured. The V1 volume is calculated using the isothermal gas transformation law.
The fuel rod is punched mechanically using a hardened steel tip. The fission gas is released into the combined fuel rod and V1 volume. The fission gas composition is analysed by mass spectrometer. The remaining fission gas is flushed back into the hot cell and the system is purged with helium. The fuel rod internal void volume is determined by subtracting the V1 volume from the combined fuel rod and V1 volume. The pressure of fission gas inside the fuel rod is calculated using the isothermal gas transformation law. For the fuel rod shown in Fig. 6, the internal void volume is 1.73 cm^3 and the fission gas pressure is 482 kPa. The accuracy is ±5 %.

A – Fuel rod
V_1 – Unknown volume (R_2 manual valve being closed)
V_2 – Standard volume (delimited by R_1 and R_2 manual valves)
S_1, S_2 – Pressure transducers

Optical microscopy

A LEITZ MM5RT optical microscope having a magnification up to x500 is used for macrographic and micro-structural analysis of irradiated fuel rod samples and for microhardness measurements. It is equipped with an image analyzer and a Vickers microhardness tester (maximum load 2 N). A computerized analysis system is used for the quantitative determination of structural features, such as grain and pore size distribution. The preparation of the samples includes precise cutting, vacuum resin impregnation, sample mounting with epoxy resin in an acrylic resin cup, mechanical grinding and polishing, chemical etching.

A macrography (magnification: x5 ÷ x10) of the as-polished sample is taken. Fig. 10 shows the macrography in the cross section of a CANDU fuel rod irradiated in the INR TRIGA reactor in a overpower test. This macrography shows a central hole caused by centre line fuel melting.

A chemical etching is applied to the fuel using a mixture of sulfuric acid, hydrogen peroxide and water to reveal the fuel microstructure and its features such as fuel restructuring. Micrographies (magnification: x100 ÷ x500) are taken from the center, middle and periphery of the sample. Fig. 11 shows columnar grains at centre of the fuel and Fig. 12 shows grown equiaxial grains with metallic fission products at middle of the fuel. The size of the equiaxial grains shown in Fig. 12 is 23.94 ± 3.78 μm being determined by interception method.

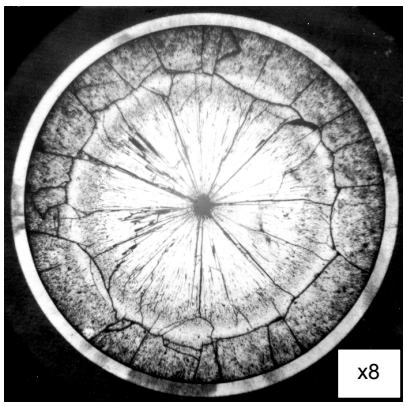


Fig. 10 Cross section macrograph of a CANDU fuel rod

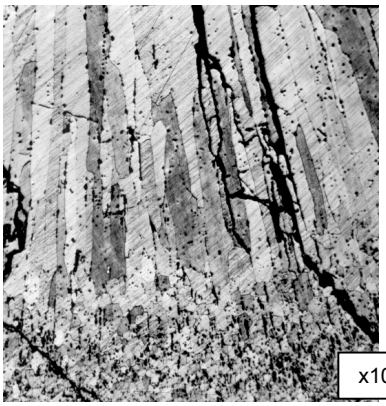


Fig. 11 Micrography at the center of the fuel

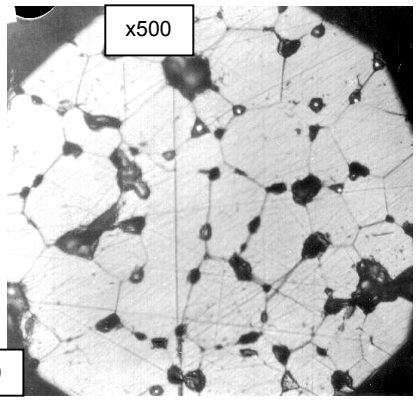


Fig. 12 Micrography at the middle of the fuel

A chemical etching is applied to the cladding using a mixture of nitric acid, lactic acid and hydrofluoric acid to reveal its hydriding (Fig. 13) and microstructure (Fig. 14). The hydride precipitations shown in Fig. 13 are orientated parallel to the sheath surfaces. A content of hydrogen of about 120 ppm was estimated by means of hydruration charts /4/. The inner and outer oxide layer thickness is measured on the micrographs taken from the as-polished sample.

The Vickers hardness measured on the surface of the sheath cross section is 240 ÷ 270 units.

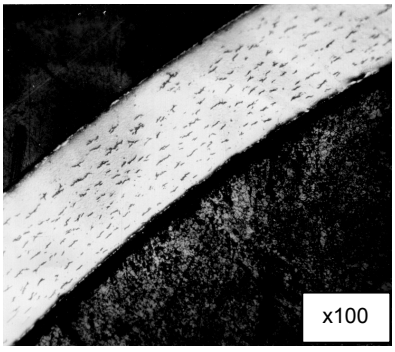


Fig. 13 Micrograph of the cladding hydriding

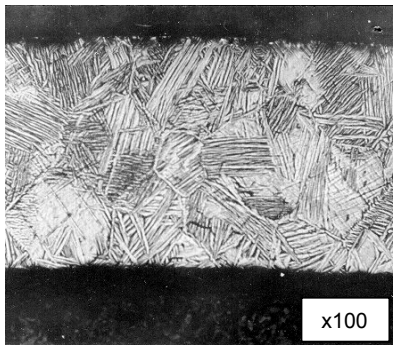


Fig. 14 Micrography of the cladding structure

Mass spectrometry

A FINNIGAN MAT 261 mass spectrometer with thermo ionisation is used to determine the nuclear fuel burnup. Two methods are used: ^{235}U depletion method and ^{148}Nd method. The procedure includes: fuel rod sample chemical dissolution in nitric acid, dilution of the fuel solution to approximately 1 gU/l, chemical separation of the U and Nd by the anion exchange chromatography, measurement by mass spectrometry of the isotopic ratios for U and Nd. The ^{235}U depletion method requires knowing the isotopic ratios $^{235}U/^{238}U$ and $^{236}U/^{238}U$ for the un-irradiated and irradiated fuel. The burnup is determined with an accuracy of ±3 %. The ^{148}Nd method is based on the determination of U and Nd concentrations in the fuel solution using the isotopic dilution technique. This technique requires a standard solution where the U and Nd concentration is well known. By this method the burnup is determined with an accuracy of ±2 %. The burnup of the fuel rod shown in Fig. 6 is 216.15 ±6.48 MWh/kgU being determined by ^{235}U depletion method.

Mechanical testing

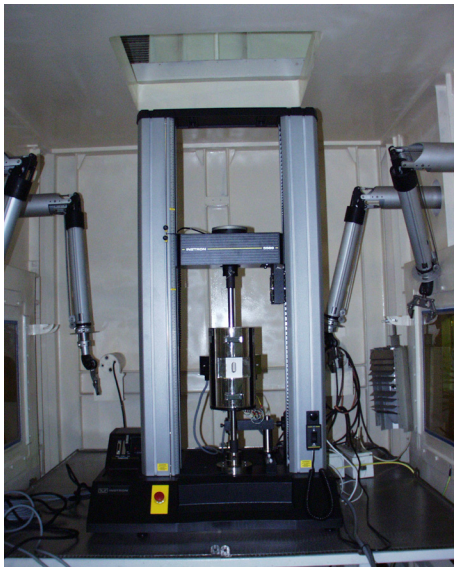


Fig. 15 50 kN INSTRON tensile-testing machine inside lead cell

The mechanical properties such as tensile, fracture strength and creep can be determined by this technique. The preparation of samples from the fuel rod is made using an ultrasonic vibrating for removal of UO_2 pellets. Two tapered end plugs are inserted at the both ends of the sample to prevent the deformation by chucking.
The old WOLPERT tensile testing machine installed into heavy concrete hot cell has been decommissioned.

A new 50 kN INSTRON 5569 tensile-testing machine has been installed into a new lead hot cell (Fig. 15). Available test conditions include a temperature range from 20°C to 1000°C under air atmosphere. Crack growth monitoring and data acquisition and handling are fully computerized. So far, only tests on the un-irradiated samples have been performed.

Conclusion

The post-irradiation examination of experimental CANDU type fuel rods manufactured by INR and tested in the TRIGA reactor has been performed at the INR PIEL since 1984. The procedures of PIE have been pursued effectively and various data of PIE have been collected and analyzed. During 20 years of post-irradiation examination, the PIEL has performed activities to provide the necessary capabilities in order to support the research and development programs for nuclear fuel and materials.

Examinations such as visual inspection and photography, dimensional measurements, eddy current testing, gamma scanning, puncture and fission gas measurement, metallography and ceramography, and burnup determination by mass spectrometry have been carried out satisfactorily. In the future, some in-cell and out-cell equipments will be provided in order to carry out the PIE on the fuel bundles discharged from Cernavoda NPP.

Bibliography

/1/. M. Pârvan, I.L. Tuturici, R. Dobrin, M. Popov, R. Rădulescu, V. Toma, The post-irradiation examination of CANDU type fuel irradiated in the Institute for Nuclear Research (INR) TRIGA reactor, Final Report for the IAEA Research Contract No.7756/R1/RB, Sept. 1995.
/2/. A. Alexa, T. Crăciunescu, G. Mateescu, R. Dobrin, The tomographic maximum entropy method in the 3-D analysis of nuclear fuel pins, Journal of Nuclear Materials, 218, 1995, 139-142.
/3/. T. Crăciunescu, R. Dobrin, I.L. Tuturici, The analysis of irradiated failed nuclear fuel rods by gamma computed tomography, Journal of Nuclear Materials, 246, 1997, 37-42.
/4/. B. Z. Hyatt, Metallographic standards for estimating hydrogen content of zircaloy-4 tubing, Report WAPD-TM-1431, 1982.