COMPARATIVE EXAMINATION OF THE FRESH AND SPENT NUCLEAR TRIGA FUEL BY NEUTRON RADIOGRAPHY

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ABSTRACT

At the Institute for Nuclear Research (INR) there is in operation an underwater (wet) neutron radiography facility (INUM) designed especially for nuclear fuel investigation. INUM was involved in CANDU experimental type and TRIGA type nuclear fuel investigations. In this paper are presented the results after investigation of the nuclear fuel TRIGA-HEU and TRIGA-LEU, fresh and spent, using transfer method with metallic foils of dysprosium and indium and radiographic films (38 cm x 10 cm). This method is the most suitable for spent fuel and offers a high geometrical resolution of the images that subsequently are digitalized with a professional scanner for films. From the images obtained for TRIGA-HEU and TRIGA-LEU with different degree of burn-up there are established the opportunities to use dysprosium or indium converter foils based on their response to thermal or epithermal neutrons to evaluate the degree of burn-up, dimensional measurements, defects etc.

Key words: transfer neutron radiography method, TRIGA-HEU, TRIGA-LEU

Introduction

Non-invasive investigation of the spent nuclear fuel by neutron radiography proved its value from the beginning of this technique in the 50s of the 20th century using transfer method with metallic foils (usually dysprosium and indium with high neutron capture cross sections available as large area foils of uniform thickness and good surface) as neutron converters to ionizing radiations (β and γ radiations) and radiographic films (single-sided or double-sided) as image recorders. Metallic foils are kept to tens of minutes behind the investigated nuclear fuel elements in a neutron beam offered by a nuclear research reactor and in short time after activation period are put in close contact with radiographic films, usually in vacuum chambers, about five $T_{1/2}$ to transfer the radioactivity profile of the metallic foil as a latent image in the silver halide of the radiographic film revealed further by chemical developing processes [1, 2]. This technique was implemented in INR at the end of 80s after construction of the underwater neutron radiography facility. First investigations were made for experimental CANDU fuel elements shortly followed by investigations of TRIGA-HEU and TRIGA-LEU nuclear fuel elements [3, 4]. The results are useful in the perspective to evaluate the behaviour to irradiation tests of the nuclear TRIGA-LEU fuel that started to be fabricated in INR.

Experimental set-up

The underwater neutron radiography facility (INUM) is placed in the pool of the dual core TRIGA reactor, Steady State Reactor (SSR) and Annular Core Pulsing Reactor (ACPR), from INR Pitesti. ACPR is the source of neutrons for INUM. In Image 1 it is shown a picture of the INUM facility.

A special sample holder or a TRIGA cassette can be introduced in the irradiation chamber of the facility. The sample holder has three positions and can support varied fuel elements (CANDU, TRIGA and experimental fuel elements), the image quality indicator (IQI) and standards for dimensional measurements (Image 2). IQI is a standard object manufactured in INR similar to ASTM ones and has two parts, a beam purity indicator (BPI) and a sensitivity indicator (SI) like ASTM type A.

BPI offers quantitative information about neutron beam and parameters of the facility that are important for the quality of the images and SI offers qualitative information about the ability of the facility to put in evidence some details (contrast sensitivity). For this SI were defined 12 levels of sensitivity according to 12 ratios of the adjoining thicknesses (Table 1). The smaller ratio of the two adjoining thicknesses establishes the level of contrast sensitivity. Good contrast sensitivity is for level 10 and very good for levels 11-12.

The main geometrical and neutron beam parameters resulted from construction of the INUM are shown in Table 2.

A dysprosium foil and an indium foil with useful areas of 380 mm x 95 mm are used at INUM (Image 3). Both foils are fixed on an aluminium holder above a cadmium plate with 2 mm thickness. Dysprosium has a thickness of 0.025 mm and indium has a thickness of 0.250 mm. Dysprosium offers better geometrical resolution than indium but indium offers a better contrast because of its resonance at 1.46 eV capturing epithermal neutrons from neutron beam that have a better penetrability in nuclear fuel. Indium is useful with its thermal neutron absorption coefficient of 0.73 mm^{-1} for ^{Nat}In (σ_{th}= 202 b for ^{115}In) and resonance integral of 3275 b for ^{115}In.

Its activated isotope ^{116}In has a half-life of 54 min and emits β^- with maximum energy of 1 Mev and γ of 417, 1097 and 1294 KeV. Dysprosium has a thermal neutron absorption coefficient of 3.01 mm^{-1} for ^{Nat}Dy (σ_{th} =2750 b for ^{164}Dy). Its activated isotope ^{165}Dy has a half-life of 2.33 h and emits β^- with maximum energy of 1.28 Mev and γ of 95 and 362 KeV.

Fig. 1 – *Placement of the INUM on the bottom of the pool with the tip of the collimator near the core of the ACPR in the maximum thermal neutron flux*

Fig. 2 - *Special holder for nuclear fuel elements with IQI (in middle) and two other standards*

Table 1 – *Levels of sensitivity for SI*

Sensitivity	1	2	3	4	5	6	7	8	9	10	11	12
Ratio of two adjoining thicknesses	1,96	1,92	1,86	1,76	1,62	1,44	1,38	1,24	1,14	1,07	1,04	1,02

Table 2 – *Main parameters of the underwater neutron radiography facility*

Parameter	**Value**
Entrance aperture diameter of the collimator, D	7.6 mm
Collimator ratio, L/D	250
Thermal neutron flux at the tip of the collimator, Φ_{th} (gold neutron activation)	1.121 x 10^{12} n/cm^2/s
Thermal neutron intensity in image plane, I_{th} (Dy neutron activation at the tip of the collimator and in the image plane)	5.433 x 10^6 n/cm^2/s
Cd ratio of the neutron intensity at the tip of the collimator, R_{Cd}	3.41
Exit aperture of the collimator	385 mm x 100 mm
Geometrical unsharpness	0.04 mm*, 0.06 mm**
Image enlargement for an object with 385 mm height	0.49 %* , 0.74 %**

* for TRIGA cassette as holder, **for special holder

Holders (404 mm x 118 mm x 8 mm) with neutron metallic converters are placed behind investigated samples with the help of a carriage (Image 4) manipulated with a rope. The carriage has a milled slit to insert the holder with metallic converter. The activation time/experiment is about 30 minutes.

Fig. 3 - Holders for dysprosium foil (top)
and indium foil (bottom)

Fig. 4 - Carriage of the metallic foil holder

Holders with metallic neutron converters together with the radiographic film are inserted in vacuum chambers to have a tight contact between metallic foil and film to improve geometrical resolution (Image 5). These new equipment consisting in new more rigid holders and vacuum chambers offered the possibility to obtain images with better quality than previously with thinner wavy holders and transfer on table with lead weights.

The transfer of the image from converter foils to film is usually over night but would be enough five hours for indium and twelve hours for dysprosium.

Further the developed and dry radiographic films are scanned with a professional scanner (Image 6) that offers high quality digitized images used for processing and analysis with proper software for qualitative examinations and quantitative measurements. This scanner has maximum optical resolution of 2400 dots per inch (29280 x 41280 pixels for A3 size scanning area) and is proper for scanning radiographic films with optical density up to 3.8 D_{max}, offering a broad dynamic range with excellent shadow details.

Fig. 5 - Vacuum chambers for image transfer from metallic foils to radiographic films

Fig. 6 - EPSON EXPRESSION 10000XL professional scanner for radiographic films

Experiments

For experiments were used ACPR as neutron source operated in steady state at a power of 100 KW and INUM facility for neutron radiography by transfer method with dysprosium and indium neutron converter screens.

First experiment was accomplished with the special holder in which were placed: a dimensional standard with cadmium and indium stripes with well known lengths on an aluminum pin manufactured with external shape of a real TRIGA nuclear fuel, IQI and a fresh TRIGA-LEU nuclear fuel element. This experimental assembly was loaded in exposure chamber of the INUS and kept dry there for image acquisition with dysprosium (32 minutes) and indium (30 minutes) neutron converters. By registering the images of the IQI with dysprosium and indium it is possible to characterize the possibilities of selective use of these two neutron converters for further applications and especially for nuclear fuel control.

Second experiment used a TRIGA-SSR cassette as a holder for five samples: two TRIGA-LEU nuclear fuel elements, one fresh and one irradiated, two TRIGA-HEU nuclear fuel elements, one fresh and one irradiated and the dimensional standard with the same construction as a real nuclear fuel element but instead of real pellets has iron pellets with well known dimensions. Using the cassette, all investigated nuclear fuel elements are placed at the same distance from neutron converter screen facilitating accurate results at dimensional measurements when it is used the dimensional standard with iron pellets. The cassette was introduced in the same place as the special holder using a special catcher that accommodates the top of the cassette in the manner of the tool that manipulates TRIGA cassettes in the pool and kept dry there for image acquisition with dysprosium (35 minutes) and indium (30 minutes) neutron converters. By simultaneous investigation of four TRIGA nuclear fuel elements, high and low enriched in U-235, fresh or spent was intended to put in evidence the differences seen in their images by using two different neutron converters screens, dysprosium and indium. Dysprosium foil with its 0.025 mm thickness absorbs 7.25% of the incident thermal neutron beam and with 0.100 mm as it is recommended in the literature would absorb 26% of the incident thermal neutron beam. Indium foil with 0.250 mm absorbs 16.68% from incident thermal neutron beam but absorbs ~95% from incident epithermal neutron beam. The neutron epithermal component estimated to be in the image plane $I_{epi} = 2.24 \times 10^6$ n/cm^2/s (as $I_{th}/(R_{Cd}-1)$) R_{Cd}) has the main contribution at image formation using indium as neutron converter foil because of the much higher cross section for interaction by capture with epithermal neutrons than that with thermal neutrons.

After loading the INUM with samples water is evacuated from exposure chamber and it is necessary to dry every water drop inside this chamber with continuous air circulation at least 10 hours for fresh fuel.
Every neutron converter exposure was followed by an image transfer on radiographic film over night, dysprosium in a vacuum chamber and indium in the second one. Vacuum chambers assure a transfer of latent image with high geometrical resolution because of a very tight contact over surface and eliminating air is avoided scattering of β^- radiations on air molecules. In the second day started chemical developing and drying of the double-coated radiographic films with high resolution (NG1, Agfa-Gevaert Structurix D2, Kodak Industrex M).
On radiographic films were performed optical density measurements to have an additional parameter at the characterization of the investigations done by transfer method.

Results

Results obtained from first experiment with special holder with dysprosium and indium are presented in Image 7 and Image 8 and results obtained from second experiment with TRIGA cassette as holder are presented in Image 9 and Image 10.

At a qualitative analysis:

- Images 7 and 9 obtained with dysprosium offer a better geometrical resolution than Images 8 and 10 obtained with indium for contours, clad, springs etc, the contrast being good for both types of neutron converters.
- The contrast sensitivity from the Images 7 and 8 of the SI on the radiographic films is level 8 using dysprosium and level 11 using indium. Maximum contrast sensitivity can be level 12. With dysprosium as neutron converter it is possible to obtain better geometrical resolution than with indium, its thickness being of 0.025 mm, but contrast sensitivity is poorer for strong neutron absorbing materials (with boron from SI and U-235 from nuclear fuel, for example). With indium foil it is possible to distinguish a variation of 0.125 mm, from 3.325 mm to 3.450 mm, only variation of 0.125 mm from 6.525 mm to 6.650 mm was not distinguished. The epithermal component of the neutron beam offers an advantage for contrast sensitivity because of the better penetrability than thermal neutron component through strong thermal neutron absorbing materials. Because the epithermal component is registered with indium, which has a bigger thickness than dysprosium, geometrical resolution for obtained images is poorer than using dysprosium. Unfortunately, indium is a soft metal and it is not possible to obtain thinner foils to be resistant to manipulations and on other hand the exposure time would be necessary to be increased to obtain the same optical density on radiographic films as for 0.250 mm thickness, the recommended thickness in this field.
- In Image 7 obtained with dysprosium it is not seen the hole where 1 mm thickness of cadmium is put and there is not any difference between the cadmium and structural material of the BPI, both materials absorbing strongly the thermal neutrons. Instead in Image 8 it is visible the fourth hole where cadmium is put, a proof that epithermal neutrons with energy bigger than 0.55 eV are present in the neutron beam and are registered by indium foil. This epithermal component is bigger as the tip of the collimator is closer to the core of the ACPR.
- In Image 7 in the red circle it is seen a small part missing from the pellet.

Optical density measured with MACBETH-TD-518 densitometer for radiographic films offered some conclusions:

- For dysprosium is maximum 1.46 in Image 7, maximum 1.85 in Image 9 and for that obtained with indium is maximum 1,28 in Image 7 and maximum 1.83 in Image 10, in the free area of the films. Bigger optical density would be more attractive from the point of contrast sensitivity on images.

Image 7 – For *dimensional standard, IQI and fresh TRIGA-LEU nuclear fuel image obtained with dysprosium*

Image 8 – For *dimensional standard, IQI and fresh TRIGA-LEU nuclear fuel image obtained with indium*

Image 9 – For *spent TRIGA-HEU, fresh TRIGA-HEU, standard with iron pellets, fresh TRIGA-LEU, spent TRIGA-LEU image obtained with dysprosium*

Image 10 – For *spent TRIGA-HEU, fresh TRIGA-HEU, standard with iron pellets, fresh TRIGA-LEU, spent TRIGA-LEU image obtained with indium*

- The densitometry measurements were made in free area, on nuclear fuel pellets, on IQI and the dimensional standards from Images 9 and 10 also. Were observed variations in distribution of the optical density between the two sets of experiments because of the different positions of the collimator tip near reactor core, between nuclear fuel elements with diameter of 38 mm, and different distributions of the optical density between images obtained with dysprosium and indium for the same experiment due to energetic components that are registered, explained by the competitive components of the neutron beam, thermal and epithermal neutrons. Dysprosium forms image with thermal neutrons and indium both with thermal neutrons and very effective with epithermal neutrons.

- Optical densities are bigger with 0.3 at the edge of the films than at 17 mm inside due to scattered neutrons on aluminium walls of the exposure chamber and holder of samples. Images obtained with indium are less affected by scattered neutrons.

- Images on radiographic films obtained with dysprosium have the same level of grey for fresh TRIGA-HEU and fresh TRIGA-LEU or spent TRIGA-HEU and spent TRIGA-LEU at the same degree of burn-up of 20MWd. As a consequence, dysprosium is used to obtain an image correlated with the number of U-235 nuclei. Important in the determination of U-235 nuclei from fuel structure are thermal neutrons that strongly interact with these nuclei and are registered very efficient by dysprosium itself subsequently.

- With indium, the image is the same from the point of the level of grey for the same type of fuel, TRIGA-HEU or TRIGA-LEU, irrespective of the degree of burn-up. The burn-up of U-235 does not affect the transparency of the nuclear fuel at epithermal neutrons. Due to non uniformity of the optical density on horizontal direction (about 0.1) it is not possible from this experiment to appreciate the difference of optical density between TRIGA-HEU and TRIGA-LEU, but for TRIGA-HEU the optical density is slightly smaller due to a bigger number of H nuclei in the matrix fuel-moderator.

Conclusions

The equipment used at the experiments formed by INUM facility itself (collimator, exposure chamber, dry tube for carriage of the neutron converter holders), sample holders (special holder or TRIGA cassette), flat and rigid neutron converter holders and vacuum chambers assures the best conditions for high quality results represented by reproducible images of high geometrical resolution of the TRIGA nuclear fuel elements.

Use of a TRIGA cassette as a holder for investigated TRIGA fuel elements is very advantageous: easy load and unload of the nuclear fuel elements in the pool of the TRIGA reactors with available tool avoiding transfer to hot cells as in the case of the special holder, maximum five nuclear fuel elements to investigate instead of maximum three at special holder, smaller distance with 5 mm between nuclear fuel elements and converter foils that improves geometrical unsharpness (from 0.06 mm at 0.04 mm, Table 2).

The obtained images were the base for assessment of the proper metallic neutron converter, dysprosium or indium, recommended for qualitative examinations and dimensional measurements for the characterization of the TRIGA-HEU or TRIGA-LEU nuclear fuel elements. The performance of the facility to offer premises for high quality investigations is proved with image quality indicators (IQI) investigated together or in the same conditions with nuclear fuel.

Nuclear fuel elements TRIGA-HEU or TRIGA-LEU do not present defects of structural integrity, a proof of the reliability of this type of nuclear fuel.

By investigating TRIGA nuclear fuel elements with dysprosium foils it is possible to distinguish between fresh and irradiated fuel elements because the irradiated ones are more transparent to neutrons and have a bigger optical density on radiographic films. Unfortunately, it is not possible to distinguish with dysprosium foils between TRIGA-HEU and TRIGA-LEU fuel elements. The difference between images obtained with dysprosium is from nuclei of U-235 and Er-167. With dysprosium foil, based on the better geometrical resolution, are performed dimensional measurements for nuclear fuel.

By investigating nuclear fuel elements with indium foils it is not possible to distinguish between fresh and irradiated TRIGA nuclear fuel of a certain type HEU or LEU, the optical densities being the same. In this case matters more the interaction with H and U-238 nuclei of the epithermal neutrons. It is possible in the case of a uniform neutron beam on image plane to have a small difference of the optical density to distinguish between TRIGA-HEU and TRIGA-LEU, TRIGA-HEU having a smaller optical density on its fuel pellets than TRIGA-LEU.

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