ELSEVIER

Journal of Nuclear Materials 246 (1997) 37–42

journal of nuclear materials

The analysis of failed nuclear fuel rods by gamma computed tomography

Relu Dobrin [a], Teddy Craciunescu [b,*], Ioan Liviu Tuturici [a]

[a] *Institute for Nuclear Research, INR-LEPI, P.O. Box 78, Pitesti, Romania*

[b] *National Institute of Nuclear Physics and Engineering Horia Hulubei, IFIN.HH, Laboratory 8, P.O. Box MG-6, Bucharest-Magurele, Romania*

Received 19 February 1997; accepted 25 March 1997

Abstract

The failure of the cladding of an irradiated nuclear fuel rod can lead to the loss of some γ-radioactive fission products. Consequently the distribution of these fission products is altered in the cross-section of the fuel rod. The modification of the distribution, obtained by gamma computed tomography, is used to determine the integrity of the fuel cladding. The paper reports an experimental result, obtained for a CANDU-type fuel rod, irradiated in a TRIGA 14 MWth reactor. © 1997 Elsevier Science B.V.

1. Introduction

Today pressurized heavy water reactor (PHWR) fuel assemblies reach a remarkably high level of manufacturing quality and operational reliability. Therefore, many reactor cycles are completed without any fuel rod failure. However, occasional fuel rod failures occur, causing a release of fission products into the primary coolant system. Since nuclear power plants have licensing limits for the release volatile fission products to the environment (off-gas limits) detailed monitoring of the development of clad failure is necessary. Activity concentration levels near licensing limits can lead to losses in operational flexibility (load follows and in some cases may even require a reduction in plant power) [1].

During CANDU power reactor operation, the fission product concentration and the ratio between fission product nuclides in the primary coolant are the only indicators of fuel clad failures. Therefore, the only means for on-line detecting fuel clad failures are

(a) Measuring the flux of delayed neutrons (DN) emitted by the short-half-life fission products, e.g., ^{87}Br($T_{1/2}$ = 55 s), ^{137}I ($T_{1/2}$ = 24 s).

(b) Monitoring the coolant and off-gas activity concentrations, e.g., ^{131}I ($T_{1/2}$ = 8.04 d), ^{133}I ($T_{1/2}$ = 20.8 h), ^{133}Xe ($T_{1/2}$ = 5.29 d), ^{134}Cs ($T_{1/2}$ = 2.06 y) and ^{137}Cs ($T_{1/2}$ = 30.1 y).

After shutdown and fuel discharge from reactor various techniques of investigation are available to identify defective fuel assemblies and fuel rods. A number of additional techniques are being used, directly at the nuclear power plants as well as in the hot cells. Among the hot cells techniques, a powerful one is the gamma computed tomography (GCT).

At the INR Pitesti hot cell facility, several nondestructive techniques are available for the evaluation of the clad integrity. Among them the visual inspection, Eddy current control and GCT have to be mentioned. The present paper reports the performance and results obtained by using the GCT method on an experimental CANDU-type fuel rod, designed and manufactured in INR, irradiated in an 100 kW pressurized water loop of the TRIGA 14 MWth materials testing reactor, in a power ramp test.

* Corresponding author. E-mail: cteddy@roifa.ifa.ro or teddy@carmen.ifa.ro.

PII S0022-3115(97)00042-1

2. Methods

2.1. Gamma computed tomography (GCT)

GCT was used, in the last years, for non-destructive evaluation of the distribution of γ-radioactive fission products in a thin radial slice of irradiated nuclear fuel rods. The shapes of these distributions are used in experimental studies concerning topics like diffusion kinetics studies [2] or the behavior of overheated fuel in case of severe accident [3], but also in usual post-irradiation examination (see, for example, Ref. [4]). Various tomographic methods have been developed (see, for example, Refs. [4–7]).

The usual approach in GCT is to represent the two-dimensional function f, which describes the distribution of fission products in a cross-section of a fuel rod, on a rectangular grid, composed by square-shaped elements (i, j), called pixels. The activity in each pixel is considered to be constant and it is denoted by f_{ij}. The first stage in obtaining information about the distribution f is to measure the gamma emission coming from a narrow strip of the fuel rod, focused by a parallel collimator, that determines straight lines (projection rays) on the cross-section (see Fig. 1). Such a measurement in tomography is called a projection element p. A set of projection elements, obtained by successive and equidistant displacements of the collimator, constitutes a projection. Each element in a projection is denoted by the index k. Several projections can be obtained at different rotation angles of the rod. The angle is called the projection angle and it is denoted by the index m. In conclusion, each measurement p_{km} means an integral of the γ-radioactive distribution. The relationship between the two-dimensional function f and its projections p is described by the equation

$$p_{km} = \sum_{i,j} w_{ij,km} f_{ij}, \tag{1}$$

where $w_{ij,km}$ is the weighting factor with each pixel (i, j) contributing to the projection element p_{km}.

Besides the geometrical contribution, proportional to the intersection length of pixel (i, j) with the projection ray (k, m), the weighting factor includes also information about attenuation and collimator penetration.

The specific problem in the tomographic reconstruction in case of nuclear fuel rods is the small number of projections that can be measured in a reasonable time: the sample is placed in a hot cell and its radioactive content is measured via a long collimator which determines a small detection efficiency; so, a long time is necessary for the acquisition of the experimental data. Consequently, only a small number of projections can be obtained in a reasonable time. Therefore, Eq. (1) provides incomplete data. A supplementary criterion must be introduced to compensate the lack of information. Such a criterion is the maximization of the informational entropy of the system of $N \times N$ pixels:

$$-\sum_{i,j} f_{ij} \ln(f_{ij}) \tag{2}$$

taking into account the experimental data via Lagrange multipliers (Eq. (1)). The usefulness of the maximum entropy criterion for tomographic reconstruction was suggested for the first time in Ref. [8]. An implementation of this method for fuel rods analysis was reported and described in details previously [9].

2.2. Clad defects identification

As mentioned before, fuel rod failures cause a release of fission products into the primary coolant system. Fission gases accumulated in the free volume of a fuel rod escape through the clad defect. Water entering the fuel rod reacts with fission

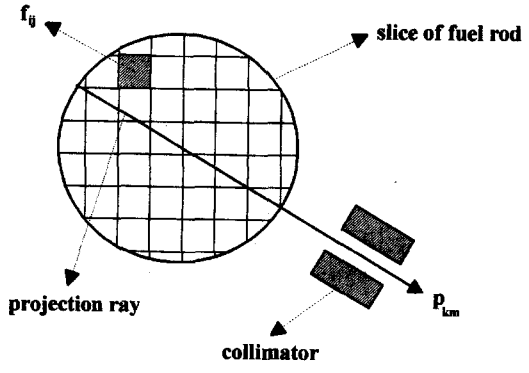

Fig. 1. The principle of tomography.

products forming volatile chemical compounds. These volatile compounds may escape in a similar manner to the fission gases, other compounds may dissolve and may be carried outside the fuel rod as dissolved species. Consequently, the distribution of these fission products, in the cross-section of the fuel rod, is modified. GCT is used here to obtain such distributions, in the area of the fuel rod suspected to be damaged. These distributions will be compared with the distributions obtained on an intact fuel rod, with the same characteristics and irradiated in the same conditions. A significant difference between the shapes of these distributions emphasizes that a failure of the cladding material occurred.

The formation of some of the γ-radioactive fission products like ^{134}Cs and ^{137}Cs is preceded by the formation of the gaseous fission products ^{133}I and ^{133}Xe in the case of ^{134}Cs:

$$^{133}_{53}\mathrm{I} \xrightarrow{\beta^- (20.80\,\mathrm{h})} {}^{133}_{54}\mathrm{Xe} \xrightarrow{\beta^- (5.27\,\mathrm{d})} {}^{133}_{55}\mathrm{Cs} \xrightarrow{(\mathrm{n},\gamma)} {}^{134}_{55}\mathrm{Cs} \xrightarrow{\beta^- (T=2.08\,\mathrm{y})} {}^{134}_{56}\mathrm{Ba} \tag{3}$$

and ^{137}I and ^{137}Xe in the case of ^{137}Cs:

$$^{137}_{53}\mathrm{I} \xrightarrow{\beta^- (T=24.0\,\mathrm{s})} {}^{137}_{54}\mathrm{Xe} \xrightarrow{\beta^- (T=3.8\,\mathrm{m})} {}^{137}_{55}\mathrm{Cs} \xrightarrow{\beta^- (T=30.0\,\mathrm{y})} {}^{137}_{56}\mathrm{Ba} \xrightarrow{\gamma (T=2.6\,\mathrm{m})} {}^{137}_{56}\mathrm{Ba}. \tag{4}$$

When the fuel cladding is fractured, a release of ^{134}Cs and ^{137}Cs or their gaseous precursors occurs, mainly because the Cs compounds are soluble in water. Consequently the distribution of ^{134}Cs and ^{137}Cs is strongly altered in the neighborhood of this region of the fuel rod. On the other hand, other fission products are not soluble in water and their formation occurs without the formation of any gaseous precursor. A typical example is ^{95}Zr that is, in addition, suitable for γ-spectrometric measurements. The distribution of ^{95}Zr is not influenced by defects of the fuel cladding. The reason is that ^{95}Zr is dissolved in the fuel (solid solution). A comparison of the distribution of ^{134}Cs or ^{137}Cs and ^{95}Zr, respectively, can be used to decide on the integrity of the fuel cladding in the suspected region. If the shapes of the distributions are similar, then it can be stated that no defects occurred. If the distributions reveal an important distinction between theirs shapes, then this is the evidence that a failure of the cladding material occurred. The method is very simple but it must be applied with caution. One must first ensure that the modification of the shape of the distributions of ^{134}Cs or ^{137}Cs was not generated by other effects like a global heating of the rod.

The similarity of the shapes of the distributions can be judged by a visual comparison of the tomographic reconstruction. However, in order to have also a quantitative evaluation of the resemblance of two distributions we used the following correlation coefficient:

$$r = \frac{N^2 \Sigma_{i,j} f_{ij}^{(a)} f_{ij}^{(b)} - \Sigma_{i,j} f_{ij}^{(a)} \Sigma_{i,j} f_{ij}^{(b)}}{\left[N^2 \Sigma_{i,j} \left(f_{ij}^{(a)}\right)^2 - \left(\Sigma_{i,j} f_{ij}^{(a)}\right)^2\right]^{1/2} \left[N^2 \Sigma_{i,j} \left(f_{ij}^{(b)}\right)^2 - \left(\Sigma_{i,j} f_{ij}^{(b)}\right)^2\right]^{1/2}}, \tag{5}$$

where $f^{(a)}$ and $f^{(b)}$ are the compared distributions. This correlation coefficient has the value 1 if $f^{(a)}$ and $f^{(b)}$ are identical.

3. Experiments and results

The method described here was used in several experiments. We report here the GCT results, obtained in the frame of the non-destructive post-irradiation examination of six experimental CANDU-type fuel rods. Their main characteristics are: the fuel material is sintered uranium dioxide with 2.5% ^{235}U enrichment, the fuel diameter is 12.15 mm, the cladding material is Zircaloy-4 with 0.4 mm thickness. The six experimental fuel rods were irradiated as an assembly in the TRIGA 14 MWth reactor core, in the existing 100 kW pressurized water irradiation loop.

In the frame of this irradiation experiment, the intention was to perform a power ramp test, characterized by the following specific operating parameters:

— fuel rod linear power, before ramp: 35 kW/m.

— ramp rate: 0.025 kW/m s.

— fuel rod linear power, after ramp: 56 kW/m.

The total time of irradiation was planned to be about 5500 h in order to obtain a burnup around the value of 195 MW h/kg U.

At the beginning of the ramp phase, performed at the end of the irradiation, a failure of the fuel cladding of one of the irradiated rods occurred. After a visual inspection four suspicious regions, located on four different rods were identified. However the conclusions of this control are uncertain, due to the fact that the surface of the rods was covered by a relatively thick layer of corrosion products. For this reason, it was decided to perform the tomographic analysis, as described in Section 2.

Tomographic reconstruction of the distribution of ^{137}Cs and ^{95}Zr was obtained for these suspicious regions. Measure-

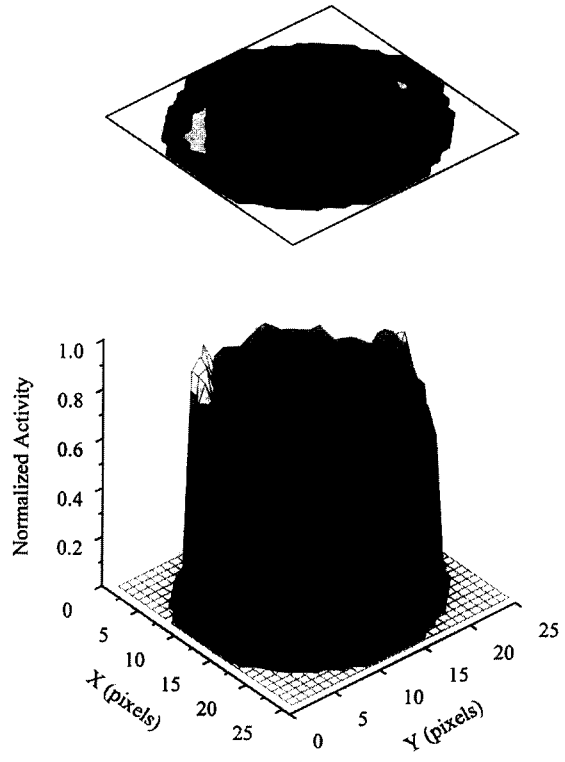

Fig. 2. Reconstruction of the ^{137}Cs (662 keV) distribution in the normal region (NR).

ments were performed using the system dedicated for usual γ-scanning tests, using a collimator slit of 0.5 mm. The entire diameter of the rod is covered by 25 relative displacements of the collimator and, in consequence, a reconstruction grid of 25 × 25 pixels is obtained. In order to minimize the total investigation time we used five equidistant projection angles. This geometry ensures a reasonable resolution of the reconstructed distributions (the global shape is relevant, not the small details). A satisfactory statistic for the ^{137}Cs (662 keV) and ^{95}Zr (724 keV) γ-photopeaks, which are recorded in parallel, was achieved for a measuring time of 60 s per projection element and a total counting time of 7200 s. The results concerning two regions are reported: one of the four suspicious regions (SR), namely one that was found to be spoiled due to the fracture of the fuel cladding and one of an intact region (IR) from an intact fuel rod. The distribution of two radionuclides is presented as follows:

- Fig. 2: reconstruction of ^{137}Cs in IR.
- Fig. 3: reconstruction of ^{95}Zr in IR.
- Fig. 4: reconstruction of ^{137}Cs in SR.
- Fig. 5: reconstruction of ^{95}Zr in SR.

Each distribution is presented, in each figure, as a 3D-surface plot (bottom) and also as a corresponding 8-level gray map (top). The correlation coefficients of the pairs of reconstructed distributions are listed in Table 1.

Table 1
The correlation coefficients between reconstruction obtained in the intact region (IR) and in the suspicious region (SR)

Distribution pair	Correlation coefficient
^{95}Zr (IR)–^{95}Zr (SR)	0.99
^{137}Cs (IR)–^{95}Zr (IR)	0.98
^{137}Cs (IR)–^{95}Zr (SR)	0.97
^{137}Cs (SR) ^{95}Zr (SR)	0.90
^{137}Cs (SR)–^{95}Zr (IR)	0.90
^{137}Cs (SR)–^{137}Cs (IR)	0.92

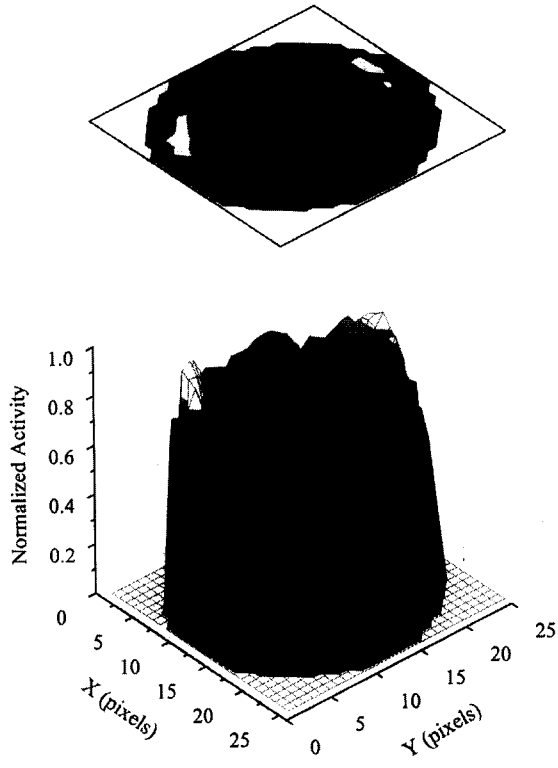

Fig. 3. Reconstruction of the ^{95}Zr (725 keV) distribution in the normal region (NR).

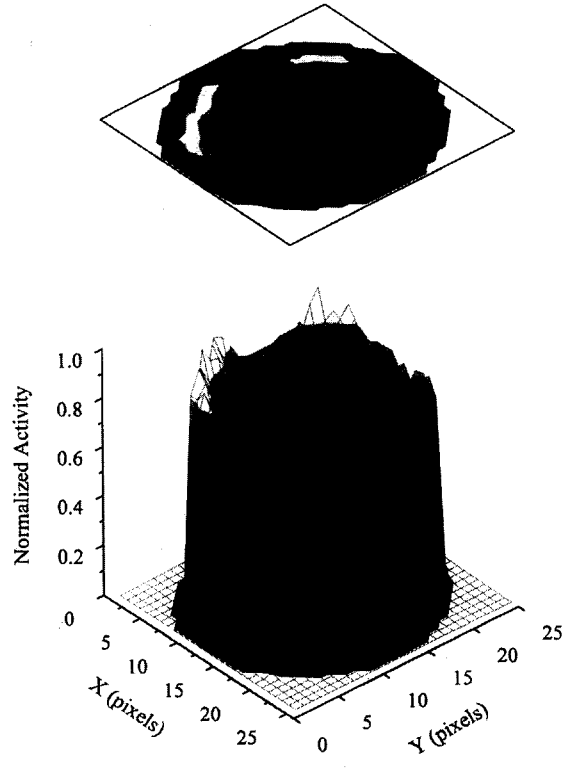

Fig. 4. Reconstruction of the ^{137}Cs (662 keV) distribution in the suspicious region (SR).

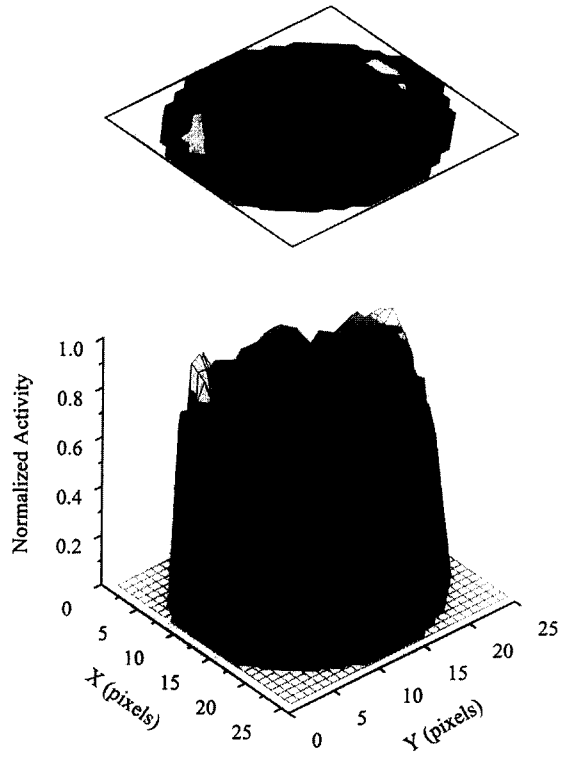

Fig. 5. Reconstruction of the ^{95}Zr (725 keV) distribution in the suspicious region (SR).

Figs. 2 and 3 reveal the likelihood of the distribution of the two radionuclides in IR. In addition the correlation coefficient (Eq. (5)) has the value 0.98. This fact ensures that a global migration of ^{137}Cs in the fuel rod, which can blur the change of the distribution did not occur. On the contrary, one can see a significant difference between the distributions of ^{137}Cs in SR on the one hand, and the distribution of ^{95}Zr in SR and also the distribution of ^{95}Zr and ^{137}Cs in IR, on the other hand. The comparative image of the distributions is emphasized by the corresponding values of the correlation coefficient. The highest value is obtained for the pair ^{95}Zr (SR)–^{95}Zr (IR). A lower but close value is obtained for the correlation coefficient in the case ^{95}Zr (SR and IR)–^{137}Cs (IR). A distinct lower value is obtained in the case of ^{137}Cs compared with the other distributions. Considering these facts we decided that this fuel rod is the damaged one. The diagnostic was confirmed later, by Eddy current control, after removing the layer of corrosion products. Similar results were obtained for the others three suspicious regions.

In conclusion we appreciate that the presented method is a powerful tool to diagnose the integrity of irradiated nuclear fuel rods. It is a full non-destructive one and provides a fast diagnostic. It can be used only if the global migration of ^{134}Cs and ^{137}Cs does not occur, but it is suitable when the surface of the fuel rod is covered by corrosion products. The examination can be performed with a usual γ-scanning system.

References

[1] IAEA Technical Report No. 322, Guidebook on Non-Destructive Examination of Water Reactor Fuel (IAEA Vienna, 1991).
[2] J. Müllauer, Untersuchungen zum Transportverhalten des Spaltproduktes Cs in UO_2-Kernbrennstoff unter Anwendung der GAMMA Computer Rep. Tomographie, Rep. GKSS 86-E-17, 1986.
[3] R.H.J. Tanke, Kema Sci. Techn. Rep. 8 (1990) 1.
[4] H.A. Buurveld, G. Dassel, Emission computed tomography on a Dodewaard mixed oxide fuel pin, ECN-C-93-065, 1993.
[5] A. Schwartz, T. Pineira, Une méthode de reconstruction bidimensionnelle de la repartition de nucleide dans un combustible nucleaire iradié a partir de scruntation gamma transversales a differentes incidences, Rep. CEA N-2272, 1980.
[6] B.K. Barnes, J.R. Phillips, M.L. Barnes, J. Nucl. Mater. 106 (1986) 147.
[7] C. Niculae, T. Craciunescu, R. Dobrin, Int. J. Energy Res. 20 (1996) 999.
[8] R. Gordon, R. Bender, G.T. Herman, J. Theoret. Biol. 29 (1970) 471.
[9] A. Alexa, T. Craciunescu, Gh. Mateescu, R. Dobrin, J. Nucl. Mater. 218 (1995) 139.