

The feasibility of express in situ measurement of the isotopic composition of uranium in fresh WWER-1000 fuel

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Abstract

A study of the feasibility and accuracy of measurement of the enrichment of fresh VVER-1000 fuel assemblies using a miniature CdZnTe probe is performed, and the possibilities of improvement of the analytical procedure are briefly discussed.

Keywords

WWER-1000 fuel, enrichment measurement, CdZnTe detectors, Monte Carlo simulation

1. Introduction

Enrichment measurement is a part of the inspection routine applied for each fresh fuel assembly delivered to a nuclear power plant. The operational environment and the test schedules impose a number of quite essential restrictions in terms of measurement duration, robustness and ease of use of the instrumentation, etc. The usual absence of dedicated calibration standards, as well as the variable enrichment profiles of the most common types of fuel assemblies pose further difficulties. Nonetheless, the manufacturer's fuel enrichment uncertainty limit of $\pm 0.05\%$ calls for a commensurable precision of measurement.

It should be noted in advance that a quite satisfactory accuracy of enrichment measurement is achievable without calibration standards through the use of a semi-planar Ge detector and the well-established and commercially available MGAU spectrum analysis software [1, 2]. However, because of the size and design of this type of detectors, they can be placed only outside the fuel assembly. Thus, in the most common case of peripheral fuel rods with lower enrichment, an additional computational procedure would be needed to resolve the contributions of fuel rods with different nominal enrichments to the aggregate spectrum, and this procedure will introduce an inevitable bias to the evaluation results.

An advantageous measurement geometry can be achieved with small detector probes which fit within the central (instrumental) tube of the fuel assembly. The most common type of detectors which meet this requirement are the CdZnTe detectors. On the other hand, their poorer energy resolution as compared with liquid nitrogen cooled Ge detectors does not allow, or at least makes extremely difficult, the application of detailed spectrum analysis methods analogous to those implemented in the MGAU software. Because of their higher energy resolution than CdZnTe detectors, a potentially better solution would be the similarly small-sized CdTe detectors with thermoelectric cooling.

The present discussion is focused on the example of a small CdZnTe detector employed at the Kozloduy NPP (model SDP 310/Z/LC/20 S by the Latvian manufacturer Ritec [3, 4]). For the purpose of enrichment measurement, the detector is inserted into the central tube of the fuel assembly. Since in this position the detector is surrounded by fuel rods of uniform nominal enrichment, and those represent the overwhelming number of fuel rods in the assembly, the measurement results have the potential of being adequately representative of the entire assembly.

2. Method

The indicator of fuel enrichment currently adopted at the Kozloduy NPP is the area of the 185.7 keV peak of ^{235}U . With the quite short measurement time of 600 s, the number of counts at this peak is in the range of 5000 – 6000. Nevertheless, from Figure 1 it can be seen that the 185.7 keV peak areas can resolve sufficiently well between the most common standard enrichments of WWER-1000 fuel rods – 3.6 %, 4.0 % and 4.4 %.

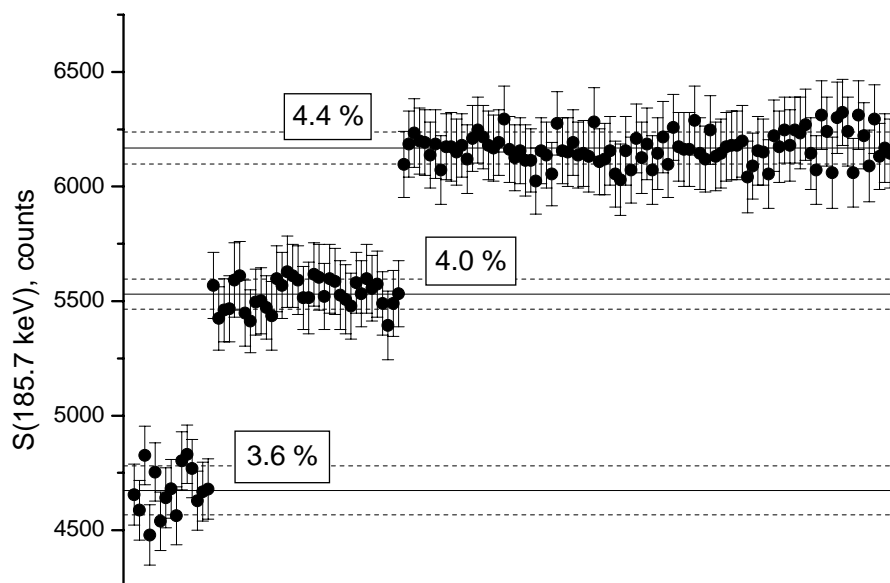


Figure 1. Counts at the 185.7 keV peak of ^{235}U . Measurement time 600 s

An improvement of this simple and straightforward approach to the relative evaluation of fuel enrichment was sought in the following directions:

- Selection of a more stable and sensitive spectral index which would accommodate for much of the effects induced by measurement geometry deviations, possible mass and density variations, instrumentation instability, etc, and thus would represent a practically absolute measure of enrichment.
- Construction of a calibration curve for the selected spectral index and evaluation of the resolution power of the measurement technique with respect to enrichment.

As an efficient aid for solving these tasks, as well as for checking the consistency of the available experimental data, Monte Carlo photon transport simulations using the MCNP Ver. 5 code [3] were performed.

The computational model is illustrated in Figure 2. This model reflects the actual design of the fuel assembly and the detector in as much detail as possible. The problem boundaries were selected through an appropriate sensitivity analysis. The response estimator is of the ‘pulse height tally’ type.

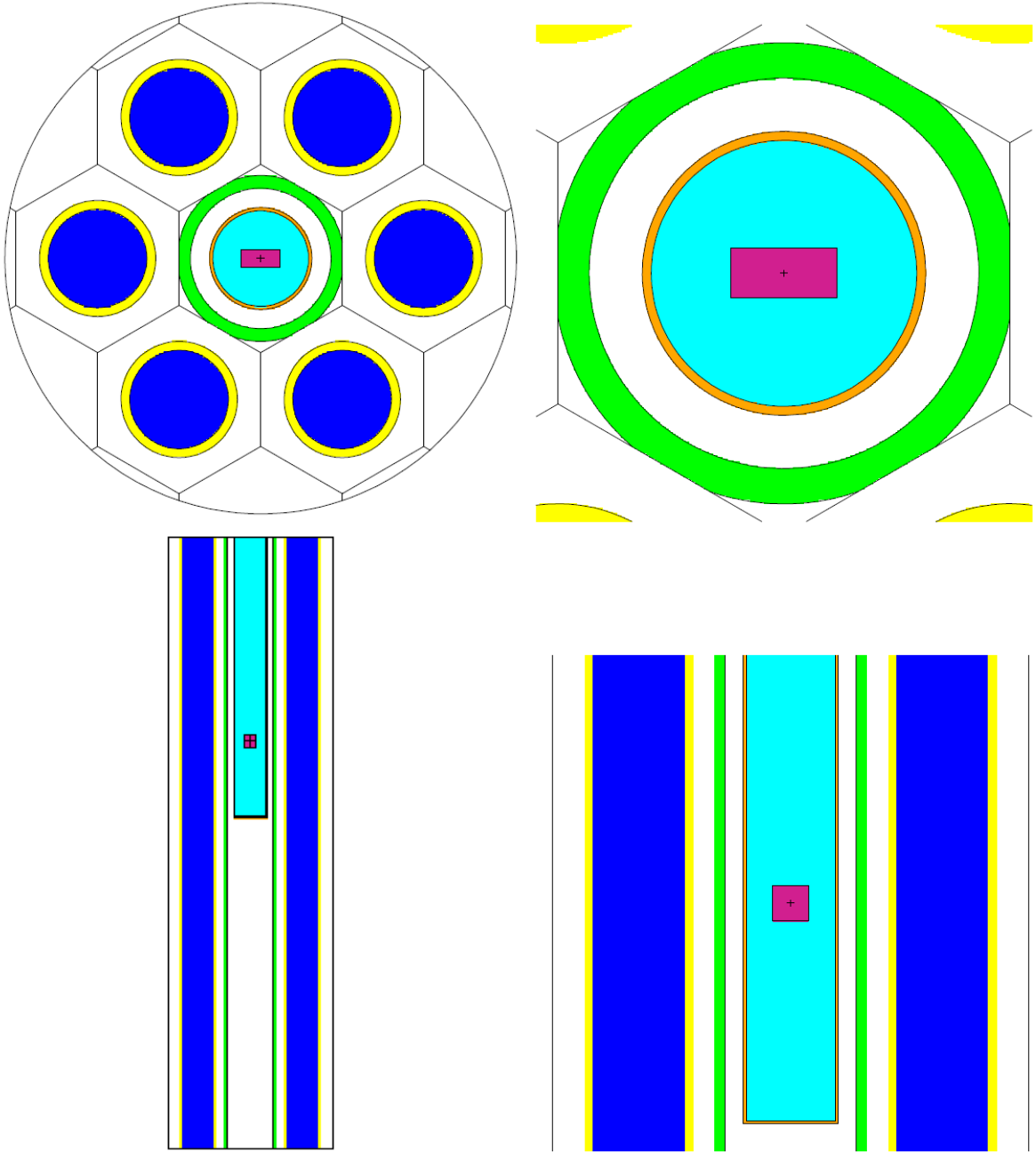


Figure 2. Problem layout for the detector and surrounding components. Horizontal and vertical sections with enlarged views

Because of the scarcity of information about the detector design, the detector model was derived from not quite reliable and mutually consistent data retrieved from different sources.

Thus, the detector is represented by a slab with dimensions $3 \times 3 \times 1.4$ mm [4]. The mass proportion between the constituent elements is assumed to be $0.9 \times \text{Cd} + 0.1 \times \text{Zn} + 1.0 \times \text{Te}$ [6]. The stoichiometric estimate of the detector material density is 5.899 g/cm^3 , whereas the density declared in [6] is 5.78 g/cm^3 . The CdZnTe slab is positioned in a stainless steel tube with an outer diameter of 8 mm and wall thickness of 0.25 mm, filled with teflon [4].

The source spectrum is based on data from [7, 8].

A common plot of an averaged (over a sample of 174 assemblies) experimental spectrum of a type ‘430’ assembly (4.4 % fuel rods in the detector vicinity) and the corresponding Monte Carlo simulated spectrum is shown in Figure 3. The relative standard deviation of the MC tally at 185.7 keV is 0.78 %.

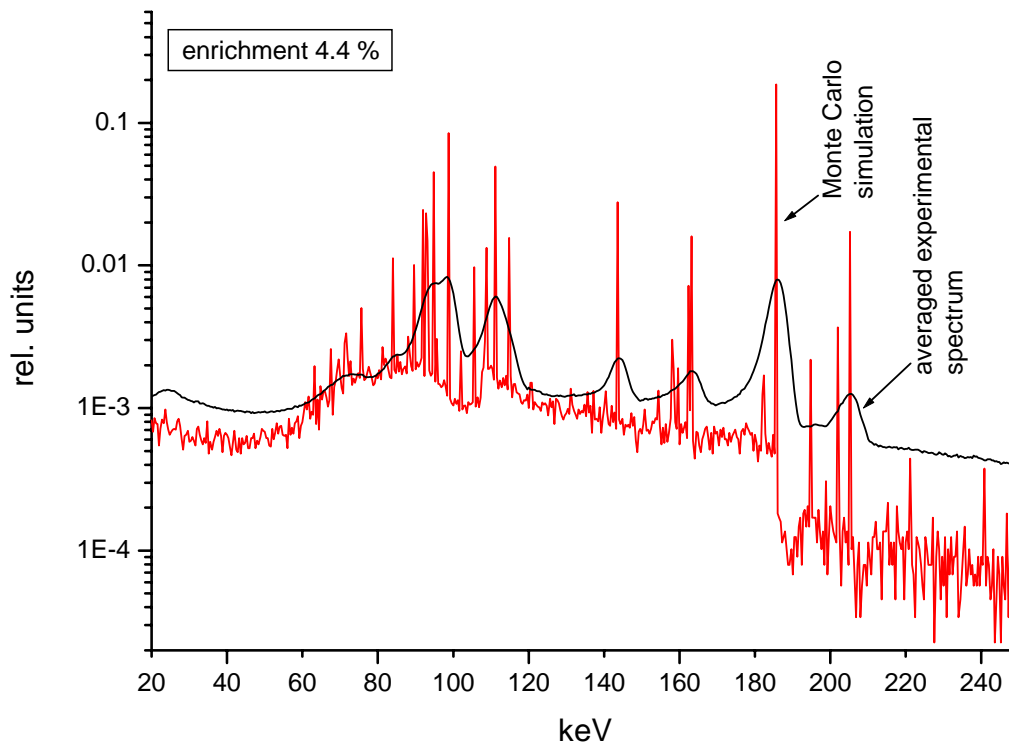


Figure 3. Experimental and modelled spectrum for a ‘430’ type WWER-1000 fuel assembly

3. Results

The stability requirement to the prospective spectral index aimed to be a measure for the fuel enrichment suggests that this index should be based on ratios or relative differences between integrals (areas) of the spectrum over selected intervals of energy.

Through studying the respective correlations, and with accounting for the poor resolution of the experimental spectra, it was established that the quantity

$$R \equiv \int_{130\text{keV}}^{215\text{keV}} S(E)dE / \int_{50\text{keV}}^{130\text{keV}} S(E)dE ,$$

where $S(E)$ is the photon spectrum, is the most sensitive practicable measure of enrichment, being as well sufficiently resistant to variations in the material properties of the assembly (including the fuel mass and density) and to fluctuations of the measurement conditions.

The energy interval [130 keV ÷ 215 keV] contains mostly lines of the spectra of ^{235}U and daughters, whereas the [50 keV ÷ 130 keV] interval contains mostly the characteristic X-ray lines of uranium.

A comparison between the modelled and the experimental dependence of R on enrichment is shown in Figure 4.

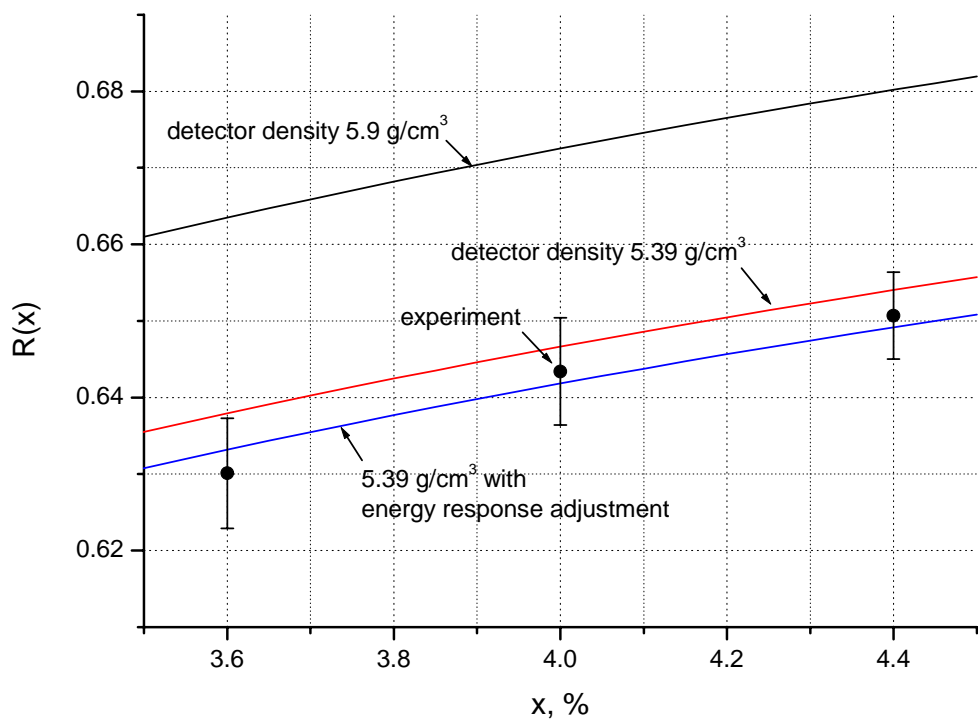
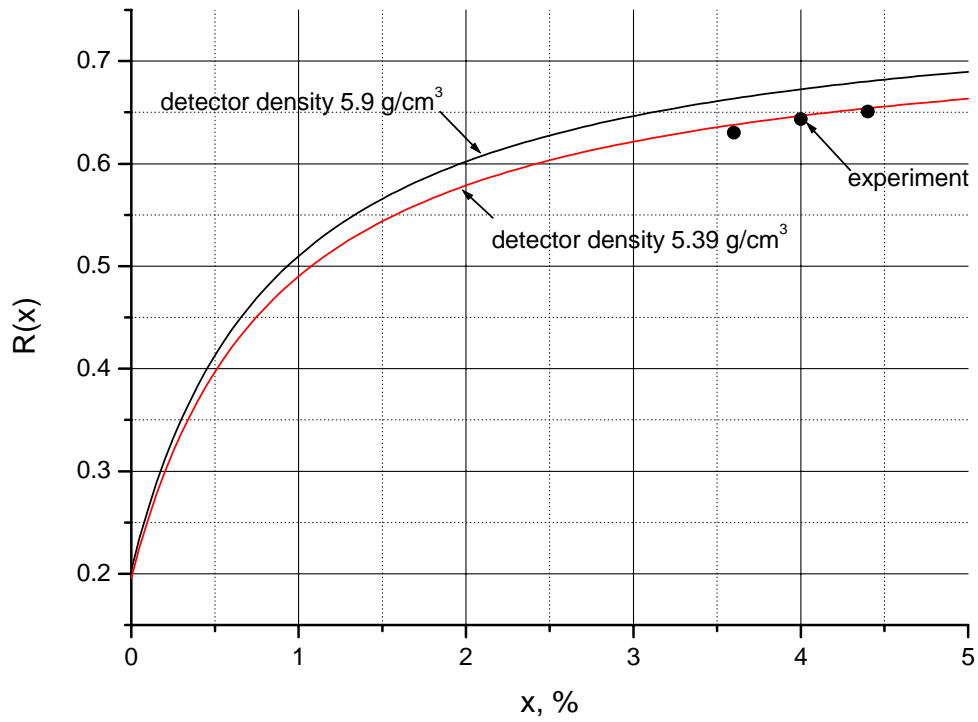


Figure 4. Dependence of the spectral index R on enrichment

In order to compensate for the various uncertainties in the computational model (especially for that of the actual sensitive volume of the detector), the detector density was adjusted to a value of 5.39 g/cm^3 which ensures a fair fit to the more reliable experimental data points at 4.0 % and 4.4 %. As seen from Figure 4, this *ad hoc* adjustment has a negligible effect on the shape and slope of the function $R(x)$. A refinement for a better fit to all three experimental data points was made through multiplying the modelled spectra by the function $c(z) = \exp(-0.0189 \times z)$, where $z \in [0,1]$ is the normalised energy in the range $20 \div 220 \text{ keV}$. This would be needed if $R(x)$ is to be used as a calibration curve, and can, of course, be avoided through further detector density adjustment or improvement of the computational model.

The slope of $R(x)$ matches the experimental trend between the enrichment values of 4.0 % and 4.4 % and is divergent from the trend between 3.6 % and 4.0 %. The available measurements at 3.6% belong to only two fuel assemblies (cf. Figure 1), and the corresponding sample is built up through multiple measurements of these two assemblies. Because of this, and also with a view to the low sensitivity of the slope of $R(x)$ to variations of the computational model, it may be concluded that the calibration curve $R(x)$ will be sufficiently reliable at least in the vicinity of the two reference points, 4.0 % and 4.4 %.

The resolution power of the spectral index $R(x)$ with respect to enrichment x can be estimated accordingly:

$$\sigma(x) = \left| \frac{dx}{dR_{\text{model}}} \right| \times \sigma[R_{\text{exp}}(x)],$$

where $\sigma[R_{\text{exp}}(x)]$ is a suitable measure of the uncertainty of experimental determination of the index R in the vicinity of a given x .

Thus, based on the current computational results, an enrichment resolution of 0.1 % maps into a required maximum relative uncertainty of R from 0.4 % at $x = 3.6 \%$ to 0.25 % at $x = 4.4 \%$. With the present relative standard deviation of R derived from the counting statistics of the available experimental spectra, this requirement would translate into a necessary measurement duration of about 2 hours instead of the currently adopted 10 minutes.

Another way to estimate this resolution power is through the standard deviations of R estimated from the accumulated samples of experimental spectra. As can be expected, the

sample standard deviations are consistent with those derived from the counting statistics, and confirm the above conclusion about the necessary measurement duration.

More specifically, the current value of the relative sample standard deviation of R is 0.9 %, whereas the corresponding value for $S_{185.7}$ (the area at 185.7 keV) is 2.5%, which is in proof of the better stability of the spectral index R . However, in the enrichment range of interest the slope of $S_{185.7}(x)$ is steeper than that of $R(x)$ which makes the former a more sensitive measure of enrichment. With the current measurement data, the enrichment resolution power of $S_{185.7}(x)$ is about 0.13 %, as compared with approx. 0.3 % for $R(x)$.

These resolution power estimates are relative in nature and say nothing about the potential bias of the evaluated enrichment. Therefore, based on the higher stability of the index R or similarly constructed rational measures, these should be preferred, while the approach to improving the enrichment resolution power would be through the accumulation of better counting statistics.

4. Further development

With a better counting statistics, a more precise choice of the instrumentation settings, and with additional calibration measurements in the energy regions of interest, aimed at accounting for the instrumentation-specific peak shapes, more sensitive spectral indices can be selected, which at the same time fulfil the appropriate stability requirement. Hopefully, this would allow to increase the measurement precision to an extent matching the manufacturer's uncertainty limits on fuel enrichment.

References

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