Beta-Ray Induced X-Ray Spectrometry for Non-destructive Measurement of Tritium in Heavy Water of the RA Reactor

Miodrag Milošević, Milan Petrović and Ivana Maksimović

***Abstract* – Measurement of tritium high-level activity in the used reactor heavy water is very important for further activities on decommissioning of the RA shut down heavy water research reactor. This paper deals with the technique for measurements of tritium high-level activity, based on spectrometry of X-rays generated by interactions of beta particles emitted from tritium with constituent elements of surrounding materials (Beta Particles Induced X-ray Spectrometry). X-rays penetrate, in general, much deeper into materials than beta particles, increasing thus indirectly significantly the detectability of tritium. This technique has few advantages compared to other methods: intensity of X-rays does not depend on the chemical forms of tritium; detector is not contaminated by tritium; etc. A detector with thin NaI crystal and beryllium window is used as an X-rays spectrometer. Proposed measurement technique differs from existing ones by using simpler equipment and materials for the experimental setup, and also, by using extensive application of Monte Carlo simulations instead of previously prepared tritium standards to quantify the amount of tritium in the sample from the measured X-ray intensity. Application of proposed technique was given for sample with 1 ml of heavy water used in the RA nuclear reactor as coolant and moderator.**

***Index Terms* – tritium high-level activity, beta particles induced X-ray spectrometry, Monte Carlo simulation, heavy water of the RA reactor.**

I. Introduction

Since its establishment in 2009, the Public Company Nuclear Facilities of Serbia (PC NFS) has continued the majority of nuclear activities previously managed by the Vinča Institute of Nuclear Sciences. The RA heavy water research reactor operated at 6.5 MW [1], is the most complex nuclear facility under the PC NFS responsibility. This research nuclear reactor was temporary shutdown in 1984, and finally permanently shut down in 2002, after decision of the Government of the Republic Serbia to repatriate the spent nuclear fuel back to Russia. During the operation of the RA reactor, a large amount of radioactive material was created by activation or contamination in different constituents and medium materials. Department for Development and Application of Nuclear Technologies at the PC NFS had undertaken a lot of activities to provide the tools and methods for measurement and detection of all relevant radionuclides, including those ones which are hard to detect, within the RA nuclear facility [2].

One of the most important issues within the process of the radiological characterisation of the reactor RA for the decommissioning purpose is characterization of several tons of activated and contaminated heavy water (used as moderator and coolant). It is known from the past, that due to long-term use of the heavy water in the high neutron fluxes, one should expect a high tritium activity level. Various techniques and devices have been developed so far for measurements of tritium activity in the tritiated water, such as calorimetry, liquid scintillation counter (LSC), etc. [3]. However, these techniques have some constraints and limitations. As first, the tritiated water above a few tens of kBq·cm^{-3} - that is just the case of the heavy water stored in the tank of the RA research reactor - is not suitable for direct measurements by the liquid scintillation counter, what implies necessity of dilution with the ordinary water. In addition, tritiated water sample has to be mixed with organic liquid scintillator and result is producing new flammable mixed radioactive waste, which later on must be processed as a new waste.

This paper presents a slightly modified methodology introduced earlier by Matsuyama et al [4]. Although dealt with the limited resources, the presented method retains all the advantages of the original methodology, meaning that the heavy water sample is measured without any physical or chemical change (and it can be returned back into the storage tank). Methodology is based on the utilisation of the X-rays generated by interactions between beta particles emitted from tritium and constituent elements of substances, and it is called Beta Particles Induced X-ray Spectrometry [4] (BIXS). Application of proposed technique is presented for sample with 1 ml of heavy water used in the RA nuclear reactor as coolant and moderator.

II. Methodology

A new experimental setup, based on the BIXS method, was built at the Department for Development and Application of Nuclear Technologies (PC NFS) for the investigation of tritium high-level activity in heavy water samples. In this method a detectability of tritium is improved using the X-rays due to their larger range; on the other side, the downside is the fact that the conversion efficiency of beta particles to X-rays is not too high. Nevertheless, the intensity of X-rays induced by β-rays is well proportional to tritium activity in water in a wide range [4].

The main goal of this work is to provide the proper interpretation of the measured tritium data in terms of the

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specific activity (in Bq·cm^{-3}) of the sample. This interpretation was supported with the results of on the Monte Carlo simulations using the MCNP-5 code [5].

Two equal aluminium rings with thin copper foil between them was used as a lid of the specially manufactured brass vial containing 1 ml of the tritiated heavy water as shown in the figure 1 (the Cu foil was covered with additional polyethylene foil to prevent detector contamination). Low energy beta particles from tritium produce bremsstrahlung radiation in the surrounding materials, mostly in the Cu foil, which was measured with thin Canberra's NaI scintillation detector Model 1702 manufactured by Canberra (Meriden, Conectitat). Thin NaI crystal with diameter of 25 mm and thickness of 1 mm was covered with Be window. It was connected to an MCA166 multichannel analyser, manufactured by GBS Electronic GmbH (Rossedorf, Germany) that uses the WinSPEC software (GBS) [6]. The detector was placed in the lead collimator to lower influence of natural radiations, figure 2.

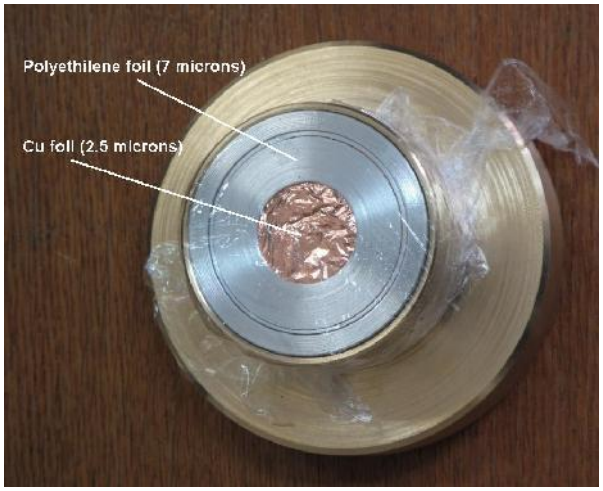

Fig. 1. Sample holder (vial), with lid (Cu foil)

Fig. 2. Experimental setup: detector and sample holder in the lead collimator

The thickness of Cu foil was determined by measurement of beta particle transmission. A tungsten shielded HP210T pancake Geiger-Mueller probe, manufactured by Thermo Electron Corporation (Santa Fe, New Mexico) was used to measure transmission of beta particles from a ^{14}C source. A geometry model of the HP210T probe, developed for the MCNP-5 code, was used to determine the response of the probe, and calculate transmission as dependence of the foil thickness. Thickness is then determined from the diagram as the value corresponding to the transmission value measured with pancake probe. Finally, the thickness of Cu foil is estimated as 2.6 μm.

The geometry model of thin NaI detector with heavy water sample, prepared for the MCNP-5 code is shown in figure 3. Both Cu and polyethylene foils are included in the model, but they are not visible separately on the drawing because they have very small thicknesses.

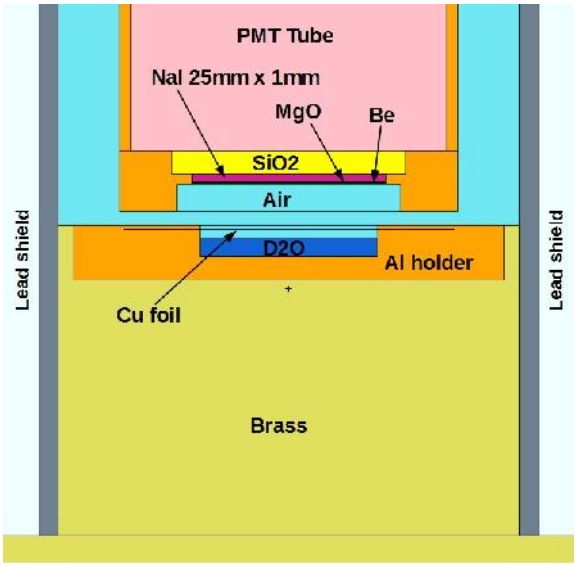

Fig. 3. Vertical cross section of NaI detector geometry model

Establishing the relation between intensity of X-rays and tritium activity in heavy water was realised in two separate steps by using the MCNP-5 code. In the first step, the X-ray leakage current through the top surface of copper foil is calculated; after that, the results of the first step was used as a source, and response of the detector, i.e., F8 Pulse Height Distribution (PHD) tally was calculated. The GEB option of the MCNP-5 code, with desired full width at half maximum (fwhm) given by

$$\mathrm{fwhm}(E) = a + b\sqrt{E(1 + c \cdot E)}\ .$$

was used to simulate Gaussian energy broadening at the energy peaks. The GEB parameters a, b, and c, equal to 0.01343, 0.07731 and 0.1436, respectively, have been chosen to reproduce the actual resolution calibration. Finally, to avoid errors due to the net area calculation, the measured pulse height distributions (PHD) and Monte Carlo simulated PHD for thin NaI detector were analysed using the ANGES software [7].

In order to validate the presented geometry model, experimentally measured pulse height distributions of X-rays were compared with the numerically built up response functions in the whole energy range of interest. The sources used in this experiment are ^{241}Am and ^{239}Pu surface disk shaped sources. The decay scheme of those radionuclides is rather complex; in the range of interest (up to ~100 keV), there are several X and γ lines. In this paper, the data from the Monograph BIMP-5, Ref. [8,9] presented in table I, were used.

TABLE I
INTENSITIES OF USED X-RAY LINES

Nuclide	Transition	Energy [keV]	Intensity [%]
^{241}Am	Lα	13.9	13.02(10)
	Lβ	17.6	18.58(13)
	Lγ	21.0	4.83(3)
	Γ	26.3446	2.40(3)
	Γ	59.5409	35.78(9)
^{239}Pu	Ll	11.62	0.1008(11)
	$Lα_1$	13.62	1.503(22)
	Lβ	17.6	2.288(23)
	Lγ	20.3	0.569(3)

The obtained measured and simulated pulse height distributions of ^{241}Am and ^{239}Pu sources for used NaI detector are given in figures 4 and 5, respectively. Comparison of the calculated and measured results shows notable differences (up to ±25%) when the single channels are considered, while these differences, when summarized for the peak area as presented in table II, decreases significantly (dropping down to less than ±4%). Discrepancies found between measured and Monte Carlo simulated pulse height distribution could stem mostly from the input values of X-rays taken from Ref. [8,9].

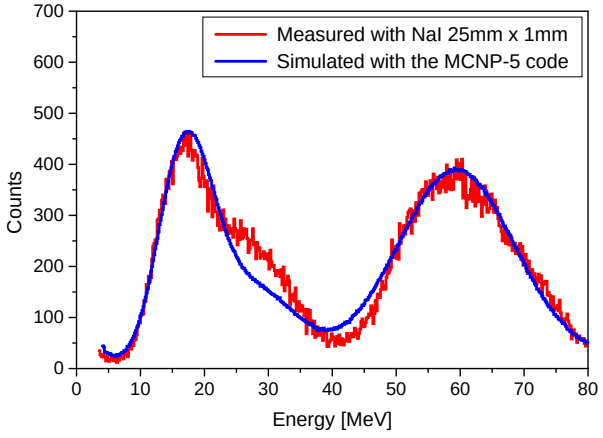

Fig. 4. Measured and simulated pulse height distributions of ^{241}Am source for thin NaI detector

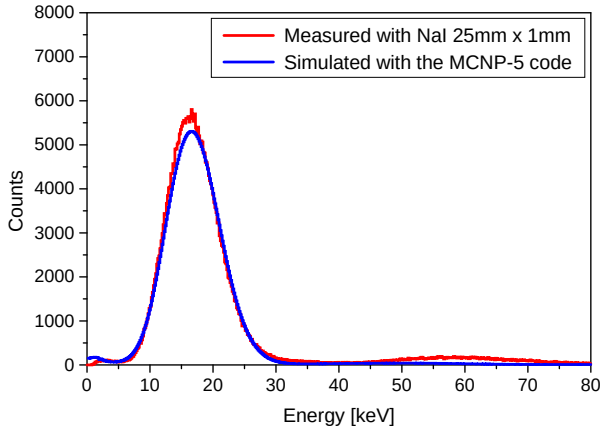

Fig. 5. Measured and simulated pulse height distributions of ^{239}Pu source for thin NaI detector

TABLE II
MEASURED AND CALCULATED EFFICIENCY OF NAI DETECTOR (CANBERRA MODEL 1702) FOR STANDARD SOURCES

Nuclide	Efficiency	
	NaI detector (Model 1702)	$100\left(\frac{\varepsilon_{cal}}{\varepsilon_{mea}}-1\right)$
^{241}Am (59.54 keV)	0.0462±0.0030	0.9
^{239}Pu (11.62÷21.3 keV)	0.0491±0.0030	3.1

III. RESULTS

Presented methodology was applied for measurement of tritium activity of heavy water used as coolant and moderator in the RA heavy water research reactor. The small volume of used heavy water (1 cm^{-3}) was poured into vial prepared according to the described model application. Pulse height distribution was recorded with NaI detector Model 1702, and also with extended range coaxial HPGe detector Model GX5020 (manufactured by Canberra), covered on the top with thin Be window. The obtained results are presented in the table III. Measured uncertainty was determined assuming that efficiency and peak area uncertainty are uncorrelated, which is case in photon spectrometry (i.e. by using the Gaussian error propagation rule). A difference less than 7% in the ^{137}Cs activity between measured results with these two detectors for the X-ray peak at 32 keV was found. Minimum measurable tritium activity for a copper coated heavy water sample equal to 10 kBq cm^{-3} was estimated by using the Monte Carlo simulation results. Finally, measurements with the thin NaI detector, and its comparison with the MCNP5 simulations using the pulse height distribution (presented in figure 6), once again confirm capability of thin NaI detector for measurements of low energy X and gamma rays.

TABLE III
ACTIVITIES MEASURED IN WATER SAMPLE

Nuclide	Activity [Bq·cm^{-3}]	
	Ge detector (GX5020)	NaI detector (ø25mm, thickness 1mm)
^{137}Cs (661.657 keV)	9.24±0.38	-
^{137}Cs (32.06 keV)	9.20±0.60	8.60±0.84
Tritium in heavy water	-	(3.18±0.26) ×10^{5}

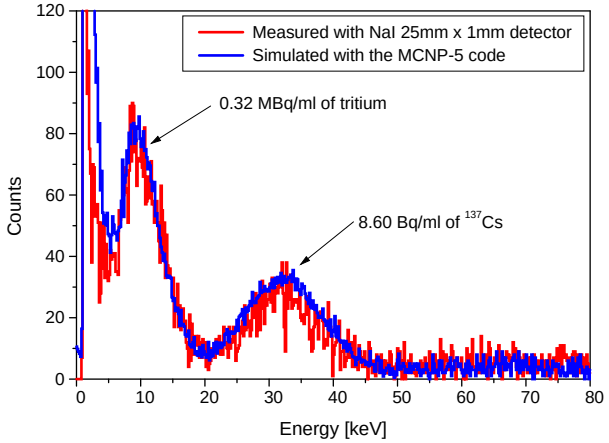

Fig. 6. Measured and simulated pulse height distributions of tritiated heavy water sample for thin NaI detector

IV. CONCLUSION

This paper demonstrates the possibility of presented methodology for fast, clean and cheap routine measuring of high-level tritium activity in water samples.

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