Measurement Using Liquid Scintillation Spectrometer-Quality Control

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***Abstract*—Liquid scintillation spectrometers (LSC) are used for radionuclide activity concentration measurements. This paper presents quality control measurements for Quantulus 1220 LSC, which is used for tritium activity determination. The quality control is verified on a monthly basis with two tritium standards. Based on these measurements two different efficiencies were calculated. Paper also presents the results obtained for tritium activity concentration in water sample within intercomparison, which are performed in Radiation and Environmental Protection Department, Vinča Institute of Nuclear Sciences. Intercomparison was organized by International Atomic Energy Agency in 2021.**

***Index Terms*—liquid scintillation spectrometer, quality control, tritium.**

I. Introduction

Liquid scintillation spectrometer is widely used for determination of low radioactivity. It can be used for detection alpha and beta radiation, Cerenkov radiation, X-rays, Auger electrons, luminescence and gamma radiation. The main advantages of using liquid scintillation spectrometer are rapidity, sensitivity, low detection limits in measurement of low energy beta emitters, such as tritium. Radioactive isotope of hydrogen, tritium ^{3}H, has low beta energy with maximum of 18 keV. Whether of natural or anthropogenic, this radionuclide is mobile in the environment, especially in water, and in biological systems. Tritium exists in three chemical forms: tritiated water (HTO), gaseous tritium (HT) an organically bound tritium (OBT). Naturally occurred tritium, produced in the upper atmosphere is oxidized to tritiated water [1,2]. Anthropogenic source of tritium are nuclear tests (between 1945. and 1963.) and nuclear facilities.

In order to determine the activity concentrations of radionuclides, optimization of detector measurement conditions should be performed. One of the basic parameters is determining efficiency. In the case of internal quality control, a periodic check of the characteristics of the detector is carried out according to the quality control plan. External quality control enables participation in intercomparisons, participation in PT schemes.

This paper presents the evaluation of the efficiency of liquid scintillation spectrometer for tritium measurement and analysis of control charts. Radiation and Environmental Protection Department, Vinča Institute of Nuclear Sciences participated in intercomparsion for tritium measurement in water sample and this paper also presents the obtained results.

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II. The Method

Measurement for the purpose of tritium activity determination in water samples are performed on liquid scintillation spectrometer Quantulus 1220. Methods for tritium determination, ASTM D 4107-08 standard method [3] and validated method [4], are accredited with the Accreditation Body of Republic of Serbia according to standard 17025.

Because of low energy of tritium, samples must be distillated to prevent the detection of other beta emitters with higher energy that could mask the tritium spectrum. One of the advantages of LSC technique is 4π geometry (Figure 1.).

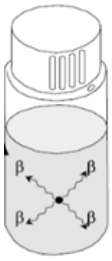

Fig. 1. 4π counting geometry [5]

Spectrometer before any measurement must be calibrated according to the calibration plan. According to the quality control plan, the efficiency check of the detector is done on a monthly basis. The value of the detector efficiency is included in the calculation for determining the activity of tritium.

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Monthly counting efficiency is determined according to method ASTM D 4107-08 Standard Test Method for Tritium in Drinking Water. Quality control is carried out with reference standard tritium solution ^{3}H 9031-OL-548/13 Czech Metrology Institute Type: ER X with activity 5.060 MBq on day 1.10.2013. and PerkinElmer Unquenched Toluene Standard 6008500 with activity 4890 Bq on day 18.11.2015. which have traceability to the BIPM.

Counting efficiency of the spectrometer, ε, is calculated using the following equation:

$$\varepsilon = \frac{R_{ST} - R_b}{A_{ST}} \qquad (1)$$

where R_{ST} is standard count rate (cps), R_b is background count rate (cps) and A_{ST} is standard activity (Bq).

Measurement uncertainty for efficiency is presented through the following equation:

$$u(\varepsilon) = \sqrt{\frac{\frac{R_{ST}}{t_{ST}} + \frac{R_b}{t_b}}{A_{ST}^2} + \varepsilon^2 \left(\frac{u(A_{ST})}{A_{ST}} \right)^2} \qquad (2)$$

where t_{ST} is standard measuring time (s), t_b is background measuring time (s).

III. Main Results

As already mentioned, according to the quality control plan, the efficiency check of the detector is done on a monthly basis, before measurement of samples.

For efficiency determination two standards are used. In addition to the standard, the background is also measured. Beside the standard from Czech Metrology Institute, for background, dead water (*DW* - tritium free water) from Miami is used, and beside the standard from PerkinElmer Background PerkinElmer is used. The first standard and *DW* are mix with 12 ml scintillation cocktail ULTIMA GOLD LLT in relation 8:12 in 20 ml polyethylene vial. Measurement time for the first standard is 300 s, and for the *DW* is 18000 s, for the second standard and for the background from PerkinElmer measurement time is 1200 s.

Figures 2. and 3. are present control chart for the efficiencies using two different standards. As a reference value, the internal calibration value is taken. Upper and lower control limits are $\pm 3\,\sigma$.

Since the LSC Quantulus 1220 has one detector and 60 positions for measurements, changing the position for the standard and for the background does not change the count rate of standard and background used in efficiency determining.

For the tritium spectrum, standard and background count rates were evaluated between channels 1 and 250 for the first standard, and for the second standard and background count rates were evaluated between channels 1 and 350.

Based on the calculated counting efficiency, the sensitivity of the instrument, *figure of merit* (*FOM*) can be calculated via following equation:

$$FOM = \frac{\varepsilon^2}{R_b} \qquad (3)$$

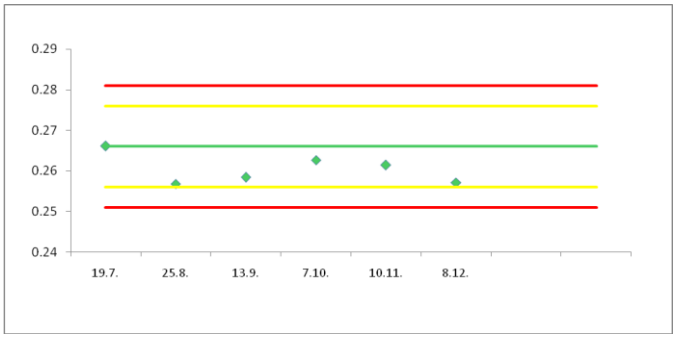

Fig. 2. Control chart for the efficiency for the standard from Czech Metrology Institute for the second half of 2021.

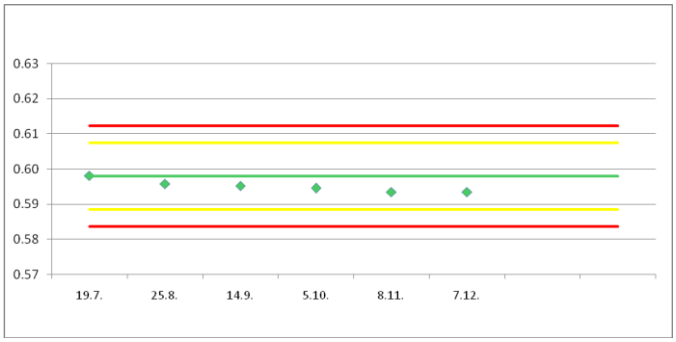

Fig. 3. Control chart for the efficiency for the standard from PerkinElmer for the second half of 2021.

Based on the results obtained for the efficiency check on monthly basis, the limits of acceptance, which is set on $\pm 2\,\sigma$, were not exceeded, observing control charts for both standards.

Using the determining efficiencies, *FOM* can be calculated based on the equation (3). For calculated efficiency for the first standard (26,6 %) and for determined background count rate (2,1 cpm), *FOM* is 337. For the second standard, for calculated efficiency for the first standard (59,8 %) and for determined background count rate (9,66 cpm), *FOM* is 370.

Radiation and Environmental Protection Department, Vinča Institute of Nuclear Sciences, National Institute of the Republic of Serbia, University of Belgrade, participated in intercopmarison ALMERA Proficiency test IAEA-TEL-2021-04. One of the required analyzes was tritium determination in water sample. For sample preparation, ASTM D 4107-08 standard method was used. The method involves preliminary distillation. An aliquot of 8 ml distillated sample is mix with 12 ml scintillation cocktail ULTIMA GOLD LLT in polyethylene vial (volume of vial is 20 ml).

Table I. presents intercomparison results. The final score is A (accepted).

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TABLE I
INTERCOMPARISON EVALUATION PT IAEA-TEL-2021-03

Sample	Reference value	Reported value	Z-score evaluation
Water	1653.6 ± 98.2	1641 ± 32	A

IV. CONCLUSION

In order to use a Liquid scintillation spectrometer to measure tritium, an analysis of control charts was performed using two tritium standards. Counting efficiencies were calculated and these values were taken as a baseline value. Monthly determination of efficiency, in accordance with the quality control plan, shows stability of the instrument. Results of intercomparison for tritium measurement in water sample show excellent values, using LSC Quantulus 1220 and accredited method.

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REFERENCES

[1] P. Calmon, J. Garnier-Laplace, “Tritium and the environment” Radionuclide fact sheet, 2010. IRSN Institut de Radioprotection et de Surete Nucleaire

[2] G. Wallova, J. Meresova, S. Zvachova, I. Petranova, I. Sykora, “New electrolytic enrichment system for tritium determination in water research institute in Bratislava and Its first results of tritium activity in precipitation” *Journal of Environmental Radioactivity* 2016 (2020) 106177

[3] ASTM D 4107-08 Standard Test Method for Tritium in Drinking Water

[4] Tecnical Procedure Note 19, Isotope Hydrology Lab., Procedure and Technique Critique for Tritium Enricment by Electrolysis at the IAEA Laboratory, IAEA 1976.

[5] I. Stojković, “Optimization of liquid scintillation spectrometer for determination of alpha and beta emitters in water” Ph.D. dissertation, Faculty of Sciences, University of Novi Sad, Department of Physics, 2015.

 ISBN 978-86-7466-930-3