Measurement 90 (2016) 412–417

Contents lists available at ScienceDirect

Measurement

journal homepage: www.elsevier.com/locate/measurement

# Designing and producing large-volume liquid gamma-ray standard sources for low radioactive pollution measurements of seawater samples by comparison between experimental and simulation results

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ARTICLE INFO

*Article history:*
Received 8 August 2015
Received in revised form 25 April 2016
Accepted 26 April 2016
Available online 27 April 2016

*Keywords:*
Portable HPGe detector
Large volume standards
Monte Carlo simulation
Efficiency calibration
Gamma spectrometry

ABSTRACT

In order to in-situ measurement of large volume water samples, using of a portable HPGe detector was considered. Because of necessity of efficiency calibration of the detector for the geometry (100 L), the large volume standard sources were prepared. Before making large volume standard sources (100 L), the Monte Carlo method has been applied in order to optimizing the calibration procedures and in agreement with experiment results, has been caused reducing the amount of produced radioactive wastes. First, the efficiency of the portable coaxial P-type HPGe detector for 1 L liquid standard sources in Marinelli beaker geometry was simulated. Then, the experimental efficiency calibration was carried out using the detector for those 1 L liquid standard sources in Marinelli beaker geometry. The detector dead layer was determined by comparison of the simulation and experimental efficiency curve results. Then, a relation between simulation and experimental measurements, that is, between pulse-height per emitted particle, F8 tally, and estimated amount of spiked radioactive solution into the 1 L distilled water in Marinelli beaker was found. Then, the efficiency calibration of the large volume liquid standard sources was simulated. The estimated amount of spiking radioactive solution into the large volume distilled water (100 L) has been taken into account dividing experimental efficiencies (in Marinelli beaker) by the simulated efficiencies (in 100 L). Finally, by spiking the large volume distilled water with the radioactive solution, efficiency calibration of the potable HPGe detector for 100 L geometry was done.


## 1. Introduction

Environmental monitoring of Persian Gulf (around the Bushehr nuclear power plant) and Oman Sea, as marine environments, is very important for experts and decision makers in Iran (not only for additional information to enhance the existing baseline data for the nation but also for worldwide database). As part of its research program, University of Isfahan was carried out research on radioactivity analysis in surface water samples of the northern coast of Oman Sea [1–3]. In the research, gamma ray spectrometry using a portable High-Purity Germanium (HPGe) detector is an important tool in marine radioactivity analysis. The energy resolution of the detector permits selective and non-destructive analysis of various radionuclides in seawater samples. As we know, radionuclides in-water can either be measured in the laboratory after collecting representative samples or measured in-field. In many studies and in our study, it is convenient to measure radioactivity concentration of seawater on-site during the field work instead of sending samples to a laboratory for analysis.

Because of the level of radioactive contamination of the marine environment is very low, one of the most effective ways for increasing the efficiency of a counting system is increasing the amount of sample [4,5]. Analysis of large volume water samples (100 L) requires prior knowledge of the full-energy peak efficiency at each photon energy for giving measurement conditions, which must be obtained by an efficiency calibration using standard

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http://dx.doi.org/10.1016/j.measurement.2016.04.065
0263-2241/

radioactive sources of very similar geometrical dimensions, density, and chemical composition of the sample under study, conditions that in many cases cannot be fulfilled [6]. Processes of designing and making the large volume standard sources are complicated and need careful and skillful technique [7,8]. In addition, the preparation must be practiced periodically, because they contain long-lived nuclides. This situation leads to an undesirable growth of radioactive wastes in the laboratory [9]. We have developed a method for designing and producing large-volume liquid gamma-ray standard sources for low radioactive pollution measurements of seawater samples by comparison between experimental and simulation results. Simulation of the calibration procedure with a validated computer code is applied as an important auxiliary tool for preparation of large volume standard sources. This simulation is very useful for both optimizing calibration procedures and in agreement with experiment results, has been caused reducing the amount of radioactive wastes produced in the process of preparation of standard sources. The present study aims in coinciding the experimental and simulated efficiency results for liquid standards in custom volumes (Marinelli beaker filled with *known*-spiked radioactive solution) and applying the results into preparation of large volume liquid standards (100 L vessel filled with *determined*-spiked radioactive solution). The large volume standards will be applied in efficiency calibration and finally in large volume liquid sample analysis. In-situ measurement of gamma ray emitting radionuclides has been carried out using a portable (HPGe) detector as a new method [2,3]. In this method, a portable HPGe detector is mounted on a polyethylene holder that is designed to keep the portable detector upright when lowered into the large volume samples. The method is very effective in environmental monitoring around the Bushehr nuclear power plant or for surveillance in an emergency situation.

## 2. Selected strategy for designing and making the large volume standard sources

Before making a large volume (100 L) liquid standard source, for both optimizing the calibration procedures and reducing the amount of produced radioactive wastes, following procedures have been applied [10–12]:

(a) The Monte Carlo method has been applied to simulate the efficiency of the portable coaxial P-type HPGe detector with energy resolution of 1.86 keV and relative efficiency of 41.3% for the line of $^{60}$Co for 1 L liquid standard sources in Marinelli beaker geometry [13–15].
(b) The experimental efficiency calibration was carried out using the detector for those 1 L liquid standard sources in Marinelli beaker geometry.
(c) The detector dead layer was determined by comparison of the simulation and experimental efficiency curve results. Then, a relation between simulation and experimental measurements, that is, between pulse-height per emitted particle, F8 tally, and estimated amount of spiked radioactive solution into the 1 L distilled water in Marinelli beaker was found.
(d) The efficiency calibration of the large volume (100 L) liquid standard source was simulated.
(e) The estimated amount of spiking radioactive solution into the large volume (100 L) distilled water has been taken into account dividing experimental efficiencies (in Marinelli beaker) by the simulated efficiencies (in 100 L).
(f) Finally, by spiking the large volume (100 L) distilled water with the radioactive solution, efficiency calibration of the potable HPGe detector for 100 L geometry was done [5].

In the present method, Regarding to known-content of radioactivity in Marinelli beaker standards, for producing of each large volume standard (with known volume), we can add accurate content of radioactive solution, straight forward in one step and without any try and error process, into large volume distilled water. The adding radioactive solution causes appropriate count rate, not high caused dead time and not low caused long time spectroscopy.

## 3. Experimental setup

Measurements were carried out using a portable coaxial P-type HPGe detector with energy resolution of 1.86 keV and relative efficiency of 41.3% from the line of $^{60}$Co. Spectrum acquisition was done using the computer software WinSPEC with a mini multichannel analyzer (MiniMCA) and spectrum analysis was done using the Fitzpeaks software. Measurements of 1 L liquid standard sources in Marinelli beaker geometries were not difficult. For large volume (100 L) configurations, the portable HPGe detector mounted on top of a 80 cm height and 46 cm diameter plastic container. A polyethylene holder is designed to keep the portable detector upright when lowered in the middle of water sample/large volume standard source. The portable detector is fitted on the holder in such a way that the center of the HPGe crystal coincided with the center of the water sample. The height of 100 L radioactive standard source/water sample in the container, with the presence of the holder, is 58 cm. Fig. 1 shows the measurement setup.

Measurements for 1 L liquid standard sources in Marinelli beakers were carried out using six liquid standard sources containing $^{241}$Am, $^{109}$Cd, $^{57}$Co, $^{133}$Ba, $^{137}$Cs and $^{60}$Co, separately [16]. The information on the standard sources is shown in Table 1. The prepared standard sources cover fully the gamma energy range of 50–1500 keV.

Fig. 2 shows six liquid standard sources, separately, in Marinelli beaker configurations for 100 L standard sources.

## 4. Simulation process

The Monte Carlo method has been applied to simulate the detection process in order to obtain spectrum peaks and to determine the efficiency curve for two modeled geometries, 1 L and 100 L [16–19]. The MCNPX code has been used for these simulations. This code is suitable for modeling the detector response, since it contains a tally, F8, which is specific for detector pulse-height determination. Thus, the detection process is simulated by the code to obtain pulse-height per emitted particle at any standard sources, that is, absolute efficiency for each spectrum peak and for two modeled geometries. The detector diagram should be inserted as an input geometry in the simulation code. In order to access the physical dimensions of the detector (Ge crystal diameter, Ge crystal length, Thickness Al window, Al cover diameter, Distance from crystal to Al window, Core diameter and Core depth) for simulation process, the detector was X-rayed [8]. In order to confirm the data corresponding to these seven physical dimensions and estimate of the detector dead layer for the next step of simulation in 100 L configuration, experimental efficiency calibration was carried out using the portable HPGe detector with 1 L liquid standard source in Marinelli beaker geometry and was compared with the simulated results in the same geometry. Marinelli beakers (Table 1) were mounted directly on top of the detector, Vice versa. The standard sources have many gamma peaks, but only 10 peaks were used in the simulations and the efficiency calculations in the experiments. So the experimental results and calculations will compare based on these peaks [20]. The particle history was chosen 9 million in the used code to reduce the pulse height tally