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Quality Control for the Quantification of Radionuclide in Radioactive Waste

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1. Introduction

Quality control (QC) procedures relate to ensuring the quality of the specific samples or batches of samples. QC may take a variety of forms including the use of; blanks, measurement standards, spiked samples, blind samples, duplicate analysis and QC samples. The use of control charts is recommended and is of great benefit, particularly for monitoring the QC control samples [1, 2]. In this study, we discuss the reliability of the analytical method to assay the radionuclide is investigated and reviewed through the measurement of the recovery of the standard materials. The background values and counting efficiencies of the radiation detection systems are measured periodically, and drawing up in a control chart for the QC. The stability of the detection systems and the reliability of the measured results are evaluated by the control charts [3]. MDA of individual radionuclide was also calculated from the background values and the counting efficiencies of the radiation detection system.

2. Experimental

2.1 Sample Pre-treatment

Open and closed vessel micro-wave acid digestion systems were used for the sample pre-treatment.

2.2 Radiation Detection System

A hyper-pure Germanium (HPGe) detection system including a main amplifier and a multi-channel analyzer are used for the analysis of the gamma-ray emitting nuclides. A gas proportional counter (GPC) system is used for the total alpha and beta counting of the radioactive waste samples. Liquid scintillation counting (LSC) system consists of a two photon amplifier tube and a coincidence coefficient circuit. LSC system was optimized for an accurate analysis of the alpha and beta radionuclide.

3. Results

3.1 Recovery of the Sample Pre-treatment

The simulated samples of dry active waste (DAW), such as latex gloves, cotton gloves, cotton socks, hats, the covers of shoes, etc., were prepared and ashed with a programmable muffle furnace. The ashes of the DAW were liquefied by using a closed vessel microwave acid digestion system. The recovery of the added metals (Cs, Fe, Ni, Re, Sr) was measured during the liquefying of the ashes of the DAW. The range of the recovery of the added metals (Cs, Fe, Ni, Re, Sr) is 97.4 - 103.1 %. The spent resins generated from a NPP were liquefied by using an open vessel microwave digestion system. The results of the recovery for the spent resin were 95.6 - 103.6 %. The recoveries of added the metal standards are entirely within the range of (100 ± 5) %, and the accuracy of them is within ± 5 %.

3.2 Recovery of the FP Separation

Fe, Sr, and Nb in the anion exchange resin were individually separated using mixed metal ion standards of Fe, Ni, Sr, Nb, Ca, Mg, Al, Zn, Cd, Cu, Zr, Mo, Pb, Mn, Ce, Co, Cr and U. After that, the recoveries of the added element standards were identified. The recoveries of them are 97.1 - 100.3 % on the whole and the accuracy is within ± 5 %.

3.3 Recovery of the VN Analysis

For recovery of ^{3}H in the radioactive wastes, ^{3}H is extracted by a leaching and distilling the standards of ^{3}H with nitric acid to remove the interference of ^{14}C and ^{129}I. After adopting ^{3}H (HTO) and removing the several interferences of the nuclides by an ion exchange method, ^{3}H became a quantitative assay. From this method, the results for the recovery of the ^{3}H standards are in the range of 97.9 - 99.7 %, and the accuracy of the recovery is within ± 5 %.

3.4 QC of the Radiation Detection System

Background value, standard samples, FWHM and the difference between the actuality peak and the library energy were measured using a gamma-ray source (^{152}Eu, 500 mL cylindrical bottles), and the control chart was drawn up from these results. The average of the background value is 1.61 cps and most of the measurement values show satisfactory results to within 2σ. The control charts for the α/β counting system are made by utilizing the standard source. The average of the background values for the alpha and beta counting systems are 0.31 cpm and 1.3 cpm, respectively and their counting efficiencies are 36.4 % for the alpha and 46.5 % for the beta measuring instrument. Using the unquenched standards (^{3}H, ^{14}C and BKG series), the background value, counting efficiency, figure of merit and the Chi-square value for ^{3}H and ^{14}C are obtained. For ^{3}H and ^{14}C, the background values are 2.52 cpm

and 3.32 cpm, and the counting efficiencies are 58.5 and 95.6 % respectively.

3.5 MDA of a Gamma-ray Spectrometer

MDA for the gamma-ray spectrometer used the method defined in NUREG 4.16 [4, 5].

$$P = \frac{2.71 + 4.66 \times \sigma_b}{LT}$$

Where P is the peak count rate and σ_b is the error in a background. For the 0.2 gram radioactive waste sample, the detection limit of ^{137}Cs is 3.2 Bq/mL while the radioactivity of the background is measured for 10,000 seconds by means of a gamma-ray spectrometer.

3.6 MDA of a Gross Alpha/Beta System

MDA of the alpha and the beta measurement system are 20.5 and 23.0 Bq/mL respectively, when measured by adopting 1 mL of a liquefied sample after dissolving 0.2 gram of it in 20 mL of an acid.

3.7 MDA of a LSC System

The background value is 2.52 cpm for ^{3}H and the counting efficiency is 58.5 %. MDA is 3.8 Bq/mL when 1 mL of a sample is counted for 50 min after dissolving 0.2 gram of the radioactive waste in 20 mL of an acid. Also, the background value of ^{14}C is 3.31 cpm and the counting efficiency is 95.6 %. In this case, MDA is 2.2 Bq/mL.

4. Conclusion

Validity of the applied analysis methods for radionuclide in radioactive wastes was examined by the reliability of the measured results of standard materials. We also selected different detectors for the analysis of the radionuclides, and composed proper counting systems. The stability of the radiation counting systems was evaluated by a periodic measurement of the background value and the counting efficiencies of the systems, and by drawing up a chart of the quality control. The data of the background values and the counting efficiencies were applied to evaluate the minimum detectable activity (MDA) of the radiation counting systems for each radionuclide. The MDAs for the gamma spectrometer and the liquid scintillation counter were calculated as 3.2 Bq/mL and 3.8 Bq/mL, respectively. The MDAs of the gross alpha/beta counting system in the alpha and beta range were 20.5 Bq/mL and 23.0 Bq/mL, respectively.

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