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Direct Measurement of γ—emitting Radionuclides in Waste Drum

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Introduction

The low—level radwaste produced from nuclear power plants, nuclear facilities, and in the process of their decommisioning is stored in waste depositary. For the safety of transport and storage of these wastes, some tests must be done. One of them is to analyse the kinds and activities of radionuclides in each waste drum. Segmented scanning gamma spectrum analysis can be used for direct measurement of gamma—emitting radionuclides in drum.[1] Gamma emitters such as Co—60, Cs—137, Ra—226, can be measured directly from outside of drum. In the case of beta emitters such as Ni—63, Nb—94, Sr—90 and transuranium alpha emitters can also be contained in drum, the destructive analysis is used for determining the compostition of radionuclides in drum, then gamma emitters are measured directly, thus, according to the composition, the activity of each radionuclide in drum can be determined. A method and system for direct measuring gamma emitters in waste drum are described in this paper, and measuring apparatus and measurement results as well.

1. Measurement principle

Assuming that the activity of a kind of radionuclide in a volume element in a drum is A_i. The drum is divided into 4 layers along its axis. A_j(j=1,2,3,4) is the activity of this kind of radionuclide in a certain layer. When the detector just faces layer k (k=1,2,3,4), the counting rate of the characteristic full—energy peak N_k can be written as:

$$N_k = \sum_{j=1}^{4} C_{k,j} \cdot A_j \tag{1}$$

where $C_{k,j}$ is the calibration factor, and is given by:

$$C_{k,j} = \int_{vj} A_i \cdot P_E \cdot D_E \cdot f_k \cdot EXP(-\sum_l \mu_l \cdot S_{l,i}) \cdot \frac{1}{r_i^2}\, dv \Big/ \int_{vj} A_i \cdot dv \tag{2}$$

Where P_E is the absolute intensity of the characteristic gamma ray with energy of E. D_E is the counting efficiency of full—energy peak for a point source in air while the distance between the point source and the center of the crystal is a unit—lenght. f_k is the incident angle correction factor of efficiency. μ_l is the linear attenuation coefficient in matter l of characteristic gamma ray of a kind of radionuclide. $S_{l,i}$ is the distance through matter l from the volume element to the center of crystal. r_i is the distance from volume element i to the center of crystal.

Besides P_E, D_E, f_θ, μ_l mentioned—above, the size of drum and its contents, geometric postion of deterction must be determined , the the calibration factor $C_{k,j}$ can be calculated by a computer. A homogeneous distribution mathematic model is adopted for calculation of calibration factor, unless the distributions of activity and medium has been determined.

2. Monitoring system

A system of directly measuring gamma emitters in drum consists of three units, detection, data collection and computation.

2.1 Detection

A Φ3" * 3" NaI (Tl) detector (Harshaw 12SS12/3A) accompanied a pre—amplifier with energy resolution 7% for 662KeV gamma ray of Cs—137.

A 10cm thick lead shadow—shield chamber of the detector, with a collimator. Another sheild chamber for sheilding external source has the same construction.

A device for drum handling, rotation and moving up and down.

2.2 Data Collection

The output from NaI (Tl) detector is amplifyed by ORTEC671—amplifier and formed and putted into ORTEC 100U—multichannel Analyzer, and then a gamma spectrum will be obtained.

2.3 Computation

All parameters and conditions are inputted. The computer (COMPAQ/25e) computes the activity of a kind of radionuclides in a drum.

3. Parameters determination

3.1 Detection efficiency

A known radioactivity point source of interested radionuclide is putted on the axis with 1m far from the center of crystal, a gamma spectrum is collected. Covell method is adopted to calculate the counting rate of characteristic full—energy peak. Certainely, the detection efficiency can be obtained. Detection efficiency of each interested radionuclide is inputted in a computer as a data file.

3.2 Incident angle correction factor of efficiency

On the point of 1m far from the center of crystal, with an angle θ of incidence, putting a point source of interested radionuclide for getting a gamma spectrum and counting rate, changing the angle θ, corresponding counting rate is measured and $f_{\theta i}$ is obtained. Each factor are also inputted as a data file.

3.3 Linear attenuation coefficient

Generaly, the waste drum is made of iron and concrete. The linear attenuation coefficient of such materials are measured. First, for a known radioactivity source outside the drum, measure the gamma spectrum of the radionuclide, then move the source off and collect gamma spectrum again, finailly move the drum off and put the same source back and collect, analyze these spectra, the linear attenuation coefficient of the waste in the drum for characteristic gamma energy of a interested radionuclide can be obtained.[2]

3.4 Direct measured of gamma emitters in drum

The waste drum is placed on a measurement device for collecting gamma spectra of different axis height, and rotated while measurement. The counting rate of characteristic full—energy peak for each radionuclide can be obtained through spectra analysis. Combined those parameters and conditions mentioned—above, the activity of each radionuclide can be computed.

4. Measurement error and minimum detectable activity

The measurement errors for activity of gamma emitters in the drum mainly come from:[2] (1) count statistical fluctuation and spectrum analysis. (2) calibration source. (3) measurement of linear attenuation coefficient. (4) waste contained in the drum. The error from the first three elements can be controlled within ±10% through increasing the masurement time , using the high resolution detector, high precision calibration source, and higher activity source for measuring the linear attenuation coefficient.

Two cases should be discussed for the errors from waste. The first is that, the distribution of medium density and activity for waste are homogeneous. For such waste, the error can be less than ±30% adopting homogeneous mathematic model for each layer. The second, for the distributions are inhomogeneous, although drum rotation and segmented scanning are used for reducing the error, the error is still higher. For a drum with the size of steel wall thickness of 4mm, height of 650mm, internal diamter of 650mm, internal concrete ($\rho = 2.26 g/cm$) covers thickness of 80mm, contained Ra—226 waste, and assuming that the distribution of solidified medium is homogeneous, the results show that , the error for inhomogeneous distribution of acitvity is ±50% (1σ). From the calculation results of different medium such as concrete, water and cotton, we can see that the error increases with the increase of medium density. For extreme inhomogeneous distribution waste, more detail scanning must be taken for deterimining the distribution, and the error may be reduced.

The minimum detectable activity of the system for Ra—226 under homogeneous distribution condition with measurement time of 1200 seconds is 5×10^4 Bq($1.4 \mu Ci$).

5. Measurement Results

5.1 simulated measuremrnt results

A cylindric plastic drum with 40cm high and 38cm diameter containing 0.1N hydrochloric acid solution of 4.13×10^5 Bq Co—60 is measured. The linear attenuation coefficient of the solution is 0.0645cm^{-1}(the results of 1.17MeV and 1.33MeV). The measured acitvity of Co—60 in the drum is

4.39E+05 Bq, with +6.3% difference compared with the known value 4.13×10^5 Bq.

5.2 Measurement results for waste drum

133 concrete soildified waste drums (200 Liter each) produced in the process of a decommisioning have been measured after being stored in sealed drum for 20 days. The results show that, the total activity of Ra−226 is 1.78×10^9 Bq, the result is 4% lower compared with reported value 1.85×10^9 Bq, Cs−137 3.09×10^5 Bq and the weight of depleted uranium is 30 kg.

One of the 133 drums, the higher activity of Ra−226 is 8.57×10^8 Bq. Detail scanning shows the Ra−226 in the drum seems to be a point source, and special measurement has been taken. The error is about ±12%. In addition, the error of each measurement for the drum is ±50% (1σ), however which mainly results from inhomogeneity. For the sum of all drums, it tends to homogeneous, so the error of total activity is less than each error. For this reason, the error of total activity for Ra−226 might be less than ±20%.

6. Conclution

6.1 For the cylindric waste drum with 650mm high and 650 diameter filled homogeneous solidified concrete, the error of activity for Ra−226 is less than ±30%. For inhomogeneous waste drum, the error is ±50% (1σ). The minimum detectable activity 5×10^4 Bq.

6.2 Simulated measurement results show a difference of +6% between the measured value and the reference for Co−60 homogeneous solution. The total activity of Ra−226 in drums is less 4% than reported value.

6.3 For a drum with higher activity, detail scanning can be taken for determining the radionuclide distribution, and then calculation of activity is carried out, which would reduce the measuerment error.

6.4 Applying Ge detector and double collimator, or other technology, the accuracy of measuring activity would be improved and the minimum detectable activity of gamma emitters whose energy is lower than 500KeV might also be reduced.

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