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CHARACTERIZATION OF RADIOACTIVE WASTE DRUMS BY NON DESTRUCTIVE GAMMA SPECTROMETRY

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Abstract - Assay of the radioactive content of waste drums is important for inventory and waste disposal purposes. The sampling of the drum content is a difficult, time and labor consuming procedure, involving special radiation protection considerations. On the other hand the characterization of waste drums containing radioactivity by measuring the drum as a whole (without opening) offers overall simplicity while minimizes labor and radiation protection requirements. In the present work a non destructive technique based on gamma spectrometry was used to assay radioactive waste in drums. For this purpose a portable NaI detector was utilized. The gamma ray detector efficiency for the volume source was derived by Monte Carlo simulations. The MCNP code was used to perform numerical simulations taking into consideration the energy of the gamma ray emitter, the matrix material, the detector efficiency, the geometric configuration employed, the size of the drum, and the wall material and thickness of the drum. The technique was verified by estimating radioactivity levels in drums containing ion exchange resin waste from the water demineralization system of GRR-1 open pool-type research reactor facility. Satisfactory agreement was observed by comparing the results of the non destructive method against analytical results of samples obtained from each drum. The simulations of the present study can easily be extended to model other materials and container types and sizes as well. Preliminary calculations for drums containing solidified radioactive sludge are presented. The technique can also be applied in other radiation protection applications such as biological radioactive waste from hospitals or other physical forms materials that can be met in radioactive waste.

Keywords: Gamma spectrometry, Monte Carlo; calibration; waste management; non destructive assay.

1 Introduction

The characterization of radioactive waste is an important obligation in waste management and safeguards. Non destructive gamma spectrometry is widely used for the characterization of drums containing radioactive waste. The technique is performed by: a) measurement of the drum as a whole [1], b) measurement of segments of the drum using scanning geometry [2,3] c) tomographic detection systems [4,5].

In the present work a 'semi-empirical' non destructive technique based on gamma spectrometry was used to assay radioactive waste in drums. For this purpose a portable NaI detector was utilized. The gamma ray detector efficiency for the volume source was derived by Monte Carlo simulations.

The technique was used to determine the activity of each isotope in drums containing ion exchange resin waste from the water demineralization system of GRR-1 open pool-type research reactor facility at NCSR "Demokritos" and now is extended to drums containing solidified radioactive sludge in cement. The distribution of source activity and density within the matrix materials was assumed homogeneous.

Satisfactory agreement was observed by comparing the results of the non destructive method to the analytical results of samples obtained from the examined drums.

2 Materials and Methods

2.1 Experimental

Two types of waste materials were examined. Ion exchange resin is styrene divinylbenzene copolymer of density 0.84 g cm^{-3} and it is used for water demineralization in co-flow regenerated units at GRR-1 pool-type research reactor facility. After the end of its useful life, the resin material is stored in 200 l iron drums for radioactivity decay and monitoring purposes, before final disposal. Radioactive sludge was solidified directly in 200 l drums by mixing with cement and water. The cement used is Ordinary Portland Cement and its composition by weight was: SiO_2: 20.35 %, Al_2O_3: 4.90 %, Fe_2O_3: 3.20 %, CaO: 62.95 %, MgO: 3.00 %, SO_3: 2.70 %, K_2O: 0.33 %, Na_2O: 0.46 %. The water to cement ratio is 0.4 by weight and the radioactive sludge in the

cement matrix is about 10%. The density of the solidified waste is 2 g cm^{-3}.

The dimensions of the storage drums are: height = 70 cm, diameter =56 cm. The drums are made of iron with wall thickness 2 mm. The storage period of drums is long enough thus short lived radionuclides had been decayed. The dose rate at the surface of the drums ranged between 1 and 4 μSv h^{-1}.

The technique was verified by estimating radioactivity levels in 25 drums containing ion exchange resins of determined activity via analytical measurements of samples obtained from each drum. Each of the 25 drums containing resins was externally monitored using Exploranium™ GR-130 miniSPEC portable gamma ray spectrometer. The GR-130 gamma ray spectrometer was equipped with a 38 mm diameter × 57 mm long NaI(Tl) scintillation detector of 7% resolution for ^{137}Cs at 662 keV. The detector was positioned at a distance of 25 cm from the drum surface at the drum mid-height level with its axis vertical to the drum's axis of symmetry. We note that the active center of the detector is considered at a depth of 4 cm from the detector surface. Three consequent measurements were performed for each drum one every 120° of drum rotation around its long axis. The duration of each measurement was 20 min. In the analysis mode GR-130 accumulates spectral data from a sample. These spectral data were transferred to a PC for analysis in terms of emitted energy level and net count contribution. Analysis was performed for ^{108m}Ag and ^{60}Co isotopes using the 433.9 keV and 1332.5 keV gamma ray emission lines, respectively. In the case that ^{137}Cs isotope is included in the waste, then the 662 keV gamma ray emission line of ^{137}Cs interferences to the ^{108m}Ag lines at 614 keV and 723 keV and they should be subtracted from the ^{137}Cs peak.

After completion of the non destructive assay, each of the 25 drums containing ion exchange resin was opened and two 200 g samples of the waste material were obtained, from different depths in the drum. The radioactivity levels of each sample were determined using gamma spectrometry. The typical procedure employed at NCSR "Demokritos" requires measurement of each sample using a calibrated NaI detector based gamma spectrometry system. The system consists of a 7.6 cm × 7.6 cm NaI detector, electronics and PC-based multi-channel analyzer. The duplicate measurements for each drum were averaged and the results were used in order to validate the results of the proposed non destructive technique.

2.2Monte Carlo simulations

Monte Carlo simulation of the drum and detector configuration was carried out using MCNP-4C2 code and cross-section data from the ENDF-VI-b library [6]. Both code and cross-section library were obtained from NEA Data Bank (France). For photons, the code accounts for photoelectric absorption, incoherent and coherent scattering, and possibility of fluorescent emission after photoelectric absorption, pair production with local emission of annihilation radiation and bremsstrahlung. MCNP computer code has been used to simulate particle transport and predict the response of sodium iodine detectors to gamma rays [7,8].

The detector was modeled as a cylinder of sodium iodide of 38 mm diameter × 57 mm length, surrounded by an aluminum layer of 1 mm thickness. The model geometry included a homogeneous distributed cylindrical volume source within a cylindrical iron drum. The wall thickness of the drum is 2 mm. Runs were performed for cylindrical sources of diameter 55.6 cm and heights ranging between 60 cm and 40 cm (thus representing different drum loads). The density was considered 0.84 g cm^{-3} for resin and 2.0 g cm^{-3} for solidified sludge in hydrated cement. The NaI crystal active center was positioned at 57 cm from the geometrical center of the drum at the drum mid-height level with its axis vertical to the drum's main axis of symmetry. To obtain the energy distribution of pulses created within the detector volume, the MCNP pulse height tally (F8) was used. Counted pulses correspond to the total energy deposited in the detector by each photon at a specified energy group. Efficiency curves in the energy range 60 to 1500 keV were predicted. A relative error of less than 5% was achieved in all simulated cases.

A normalization of the results was required in order to account for losses in the detector system, such as incomplete collection of scintillation light, quantum efficiency of the photo-multiplier tube and electronic losses that were not taken into account. To estimate these losses a relative calibration technique was employed. Measurements were performed by ^{60}Co, ^{137}Cs and ^{152}Eu standard point sources positioned at the geometrical center of an air filled drum. The results of these experimental measurements were adjusted against MNCP simulations for the same conditions. Thus, appropriate adjustment factors for the MCNP calculations were derived for given photon energies. These factors were applied for the adjustment of the MCNP derived detector efficiencies when the drum was filled with the material.

3 Results

Figure 1 shows the predicted detector efficiency as a function of photon energy for waste drums containing resin and solidified sludge in hydrated cement for two filling heights of 60 cm and 40 cm. The detector efficiency depends on the intrinsic efficiency of the sodium iodide detector, the geometrical factor, as well as the gamma ray attenuation properties of the matrix and drum wall material. The decrease in detection efficiency below 200 keV, in all cases, is due to absorption of low energy photons as they pass through the matrix material,

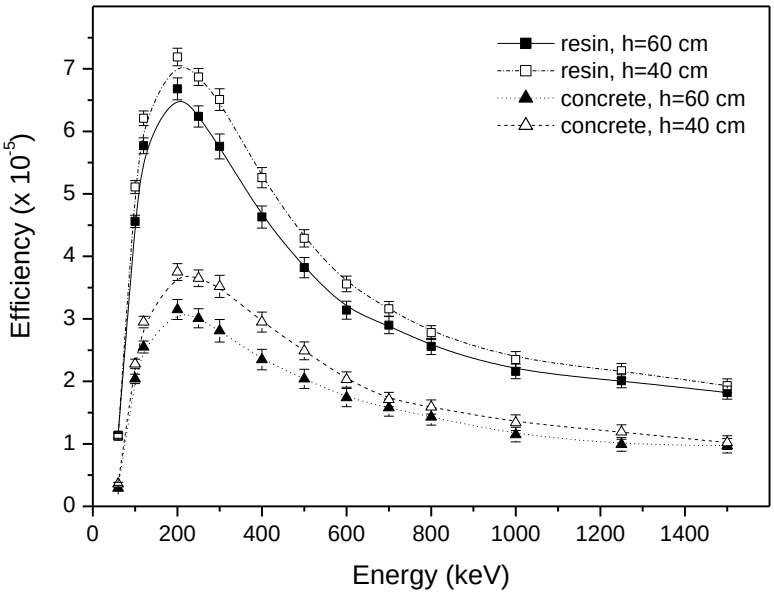

Figure 1 : MCNP predicted detector efficiency as a function of photon energy for waste drums of 60 cm and 40 cm filling heights.

drum wall and detector's mount. As it can be observed from figure 1, the efficiencies in drums containing solidified sludge in hydrated cement are significantly lower than the efficiencies for drums containing resin waste due to the higher attenuation of gamma rays in the hydrated cement. In particular, for the 60 cm filling height example, the efficiencies for the two different waste matrices differ 44% and 45% for 434 keV and 1332 keV photons, respectively. In the case of drums containing resin waste, the calculated efficiencies, for the two filling heights differed 13% and 7% for 434 keV and 1332 keV photons, respectively. For drums containing solidified sludge in hydrated cement, the efficiencies for the two heights differed 18% and 13% for 434 keV and 1332 keV photons, respectively.

Figure 2 and 3 show the activity concentrations in the resin drums as estimated by the non destructive drum assay, using efficiencies derived by the Monte Carlo method, and by the sample analysis technique, for ^{108m}Ag and ^{60}Co radioisotopes, respectively. In these figures the z-scores are also shown. The z-score is defined as $z=(c_1-c_2)/\sigma$ where σ is the combined standard deviation, $\sigma=\sqrt{\sigma_1^2+\sigma_2^2}$, of the two measurements $c_1\pm\sigma_1$ and $c_2\pm\sigma_2$ performed by the non destructive and sample analysis techniques, respectively. Thus, z-score values represent the difference between the two measuring techniques in combined standard deviation units.

Good correlation was found between the results of the two techniques for both isotopes examined (Figure 2 and 3). In particular, linear regression analysis performed on the data (95% confidence level) resulted in a linear relationship y=ax+b between the two techniques, with constant 0.03 ± 0.03 and slope 0.90 ± 0.05 for ^{108m}Ag and -0.13 ± 0.09 and 1.04 ± 0.07 for ^{60}Co, respectively. Moreover, the correlation coefficients were 0.97 and 0.95, respectively. The z-score values for both isotopes ranged between -2.7 and +1.6. The z-score mean value was -0.5 for ^{108m}Ag and -0.7 for ^{60}Co indicating an under response of the non destructive technique discussed. However, the hypothesis of no difference between the two drum assay techniques was tested in terms of a paired t-test resulting in p-values of 0.036 and 0.033 for ^{108m}Ag and ^{60}Co, respectively. Despite the size of the error introduced, the results of the non destructive technique are acceptable for such a type of waste characterization.

4 Discussions and Conclusions

A semi-empirical non destructive technique combining gamma spectrometry and Monte Carlo simulations using the MCNP code to characterize waste drums containing radioactive waste, of low level activity, was presented. The technique was verified by estimating radioactivity levels in 25 drums, containing ion exchange res in waste, and comparing the results of the non destructive method against the analytical results of samples obtained from each drum. Analysis of the results showed satisfactory agreement within 3σ between the proposed non destructive drum assay and the sample analysis techniques.

The sample analysis technique requires sampling of the drum content and therefore is a difficult, time and labor consuming procedure, involving special radiation protection considerations. On the other hand the proposed technique offers overall simplicity while minimizes labor and radiation protection requirements. Thus, it represents

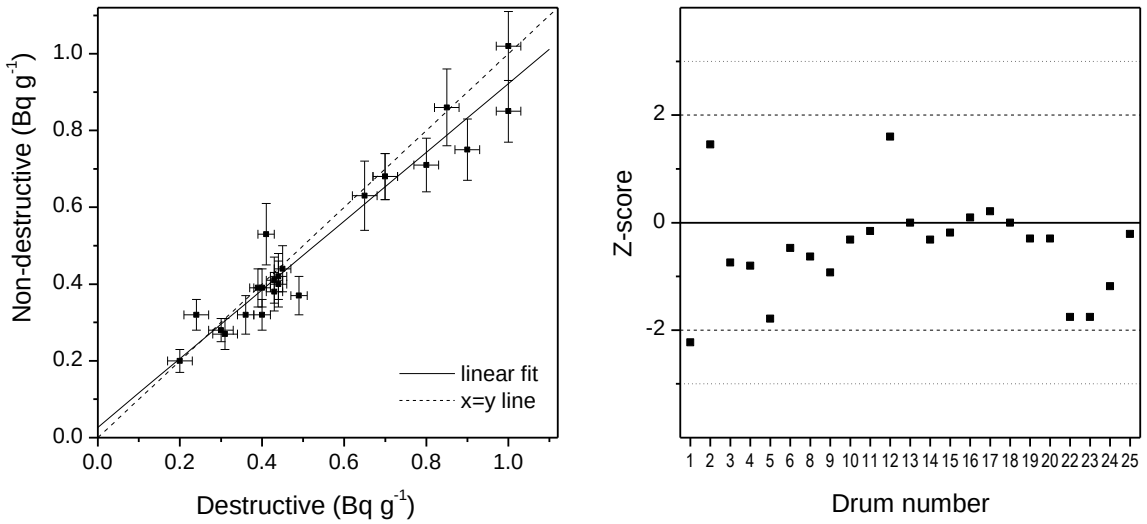

Figure 2 : ^{108m}Ag activity concentration in the waste drums as estimated by the non-destructive drum assay and determined by sample analysis.

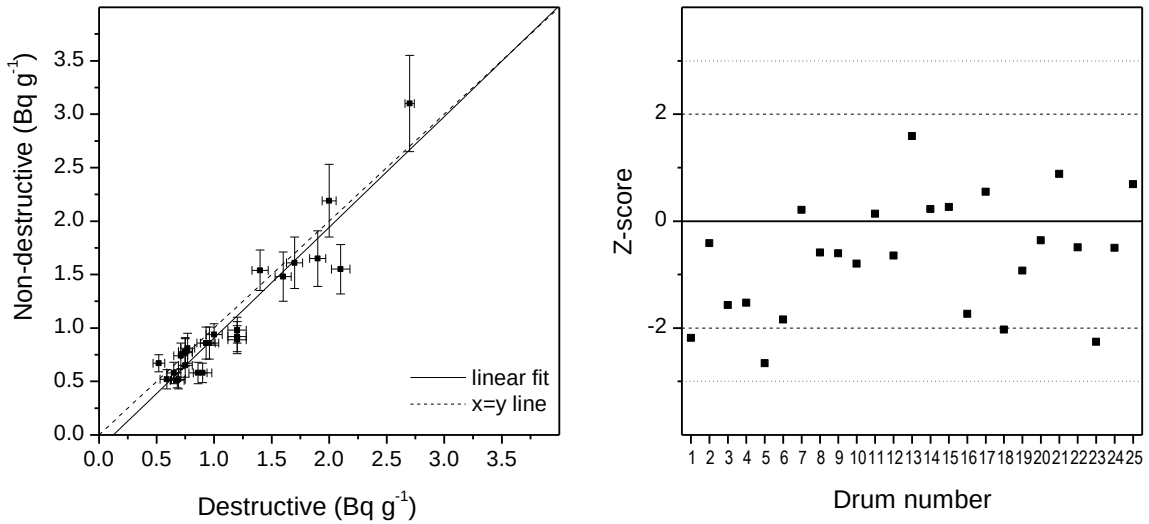

Figure 3 : ^{60}Co activity concentration in the waste drums as estimated by the non-destructive drum assay and determined by sample analysis.

a technology that can be used to assay low-activity waste drums provided the contribution from each gamma ray emitting radionuclide to the gamma ray spectrum can be resolved. The simulations of the present study can easily be extended to model other materials, container types and sizes as well.

Aknowlegments

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