Further Development in Characterization of Radioactive Waste Drums by Non Destructive Gamma Spectrometry at GRR-1

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Abstract A non destructive technique based on gamma spectrometry and application of the Monte Carlo method for detector efficiency calibration, was used to assay radioactive content waste drums. Exploranium™ GR-130 miniSPEC portable gamma ray spectrometer was used to externally monitor radioactive waste drums containing ion exchange resin waste from the water demineralization system of GRR-1 open pool-type research reactor facility at NCSR "Demokritos". Detection efficiency was predicted by Monte Carlo simulation performed with the MCNP-4C2 code and cross-section data from the ENDF-VI-b library. The code was used to perform numerical simulations taking into consideration the energy of the gamma ray emitter, the matrix material, the detector characteristics, the geometric configuration employed, the size of the drum, and the wall material and thickness of the drum. Efficiency curves in the energy range 60 to 1500 keV were predicted assuming a homogeneous distribution of the source activity within the matrix material. A relative error of less than 5% was achieved in all simulated cases. Satisfactory agreement was observed by comparing the results of the non destructive method against analytical results of samples obtained from 25drums. Moreover the MCNP model was used to examine the effect of inhomogeneities, on the accuracy of the technique and estimate the measurement bias. A technique to predict such inhomogeneities by measurements at different source to detector positions was examined.

1. INTRODUCTION

Non destructive characterization of waste drums containing radioactivity is important for inventory and disposal purposes. Two main variations of assaying techniques are used. The first employs measurement of the drum as a whole, while the second employs measurement of segments of the drum using scanning geometry or tomographic gamma ray detection systems. A review of non destructive gamma methods for waste characterization is presented in a recent IAEA publication [1].

The present work a non destructive technique to determine the radioactivity in waste drums containing ion exchange resins is discussed. The technique is based on gamma spectrometry using a portable NaI detector and Monte Carlo simulations using the MCNP computer code in order to derive the gamma ray detector efficiency for the volume source. In particular:

(i) detector efficiency was derived in a homogeneous distribution of the source activity within the matrix material

(ii) (ii) the MCNP model was used to examine the effect of inhomogeneities in activity distribution, variation of matrix material density and drum filling height on the accuracy of the technique and to estimate the measurement bias.

(iii) The results of the non destructive method were compared to the analytical results of samples obtained from 25 drums. Satisfactory agreement between the two assay techniques was observed.

2. MATERIALS AND METHODS

Experimental

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 conditioning/ disposal. 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 $4 \ \mu\text{Sv h}^{-1}$.

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 $\times$ 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. The results of the three measurements for each drum showed that the activity in the drum is symmetrical distributed around the drum axis. Analysis was performed for ^{108m}Ag and ^{60}Co isotopes using the $433.9\ \text{keV}$ and $1332.5\ \text{keV}$ gamma ray emission lines, respectively.

After completion of the non destructive assay, each of the 25 drums containing ion exchange resin was opened and two $200\ \text{g}$ 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 $7.6\ \text{cm} \times 7.6\ \text{cm}$ NaI detector, electronics and PC-based multi-channel analyzer. The duplicate measurements for each drum and the results were used in order to validate the results of the proposed non destructive technique.

Monte 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 [2]. Both code and cross-section library were obtained from NEA Data Bank (France). MCNP computer code has been used to simulate particle transport and predict the response of sodium iodine detectors to gamma rays [3,4].

In the present work the detector was modeled as a cylinder of sodium iodide of $38\ \text{mm}$ diameter $\times$ $57\ \text{mm}$ length, surrounded by an aluminum layer of $1\ \text{mm}$ 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 $55.6\ \text{cm}$ and heights ranging between $60\ \text{cm}$ and $40\ \text{cm}$ (thus representing different drum loads). The density was considered $0.84\ \text{g cm}^{-3}$ for resin. The NaI crystal active center was positioned at $57\ \text{cm}$ 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 $1500\ \text{keV}$ 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.

The effect of activity inhomogeneities on the gamma ray measurement results was examined by a set of MCNP runs, modeling extreme distributions of radioactivity in the waste drum containing ion exchange resins. Six activity distributions were modelled: (a) over the entire surface of the drum, (b) over its side cylindrical surface, (c) over one of its circular surfaces (bottom), (d) over a circular surface at the central cross-plane, (e) point source at the geometrical center of the drum and (f) line source along the main symmetry axis of the drum. Simulations were performed positioning the detector active centre at a distance of: (i) 57 cm and 32 cm from the geometrical centre of the drum and vertically to the drum's main axis of symmetry ii) 57 cm and 38 cm from the geometrical centre of the drum and on the drum's main axis of symmetry. The efficiencies ef_1, ef_2, ef_3, ef_4 were determined respectively.

3. RESULTS AND DISCUSSION

3.1 Detected efficiencies

Figure 1 shows the predicted detector efficiency as a function of photon energy for waste drums containing resin. The calculated efficiencies, for the two filling heights (40 cm and 60 cm) differed 13% and 7% for $434\ \mathrm{keV}$ and $1332\ \mathrm{keV}$ photons, respectively.

3.2 Effect of inhomogeneity

The effect of inhomogeneity and variation of matrix material density on the accuracy of the technique were discussed by Tzika et al. [5]. An increase of 10% in the resin density (from 0.84 to $0.92\ \mathrm{g\ cm^{-3}}$) in a fully loaded drum resulted in 7.2% and 9.7% decrease in detection efficiency for $434\ \mathrm{keV}$ and $1332\ \mathrm{keV}$ photons, respectively. A decrease of 10% in the resin density (from 0.84 to $0.75\ \mathrm{g\ cm^{-3}}$) resulted in 6.3% and 11.0% increase in detection efficiency for $434\ \mathrm{keV}$ and $1332\ \mathrm{keV}$ photons, respectively. The worst case of activity inhomogeneity examined was that of a line source at the center of the drum (case f) and the induced overestimation of the efficiency determination decreased from 4.50 to 1.61 for the energy range 100 to $1500\ \mathrm{keV}$. In this case the gamma rays from the source must pass through the maximum amount of the sample matrix and suffer the maximum attenuation. Therefore, the overestimation of the detector efficiency results to underestimation of activity concentration occurs. The lower underestimation of the detector efficiency with subsequent overestimation of activity concentration was that of cylindrical surface (case b). For this distribution of radioactivity and for the energy range 100 to 1500 keV , the efficiency determination increased from 0.47 to 0.70.

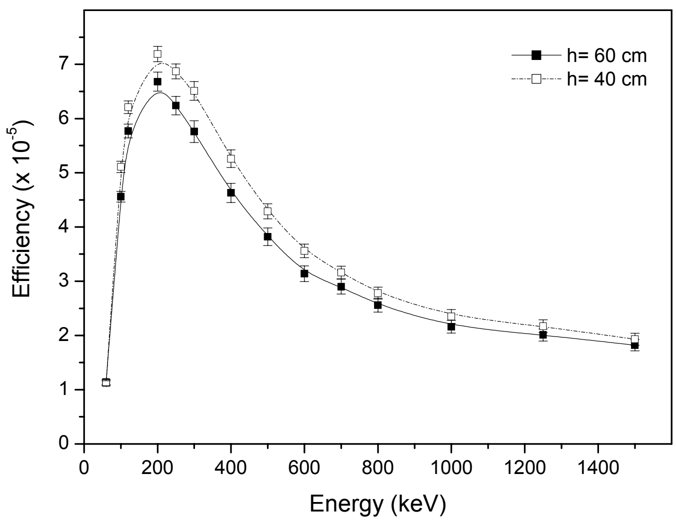

Fig. 1 MCNP predicted detector efficiency as a function of photon energy for waste drums of 60 cm and 40 cm filling heights.

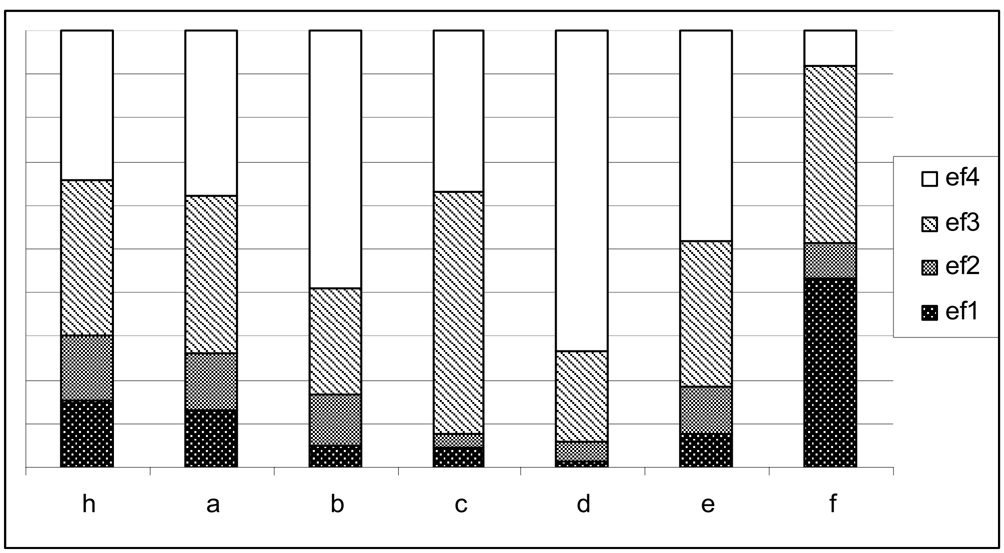

Fig. 2 The normalized predicted detector efficiencies: i) ef_1, ef_2 the detector vertical to the drum's main axis at 57 cm and 32 cm from the geometrical centre of the drum respectively ii) ef_3, ef_4 the detector on the drum's main axis of symmetry at 57 cm and 38 cm from the geometrical center of the drum respectively. The index h corresponds to homogeneous distribution and a,…, f correspond to activity distributions discussed in paragraph 2.2 .

Figure 2 shows the normalized predicted detection efficiency in four different source-to-detector positions, for each activity distribution discussed in section 2.2. Calculations were performed for 1300 keV gamma-rays, considering waste drums containing resin of 0.84 g cm^{-3} density. It is observed that there are major differences between the normalized efficiencies for different activity distributions. Deviation of the measured efficiencies to that obtained for the homogeneous activity distribution could be consider as an index of drum activity non-homogeneity.

3.2 Comparisons of Techniques

Figure 3 and 4 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 (Fig. 3 and 4). 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. 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 problem for non destructive determination of radioactive material content in a drum is difficult. Even if the attenuation coefficients and the geometrical parameters of

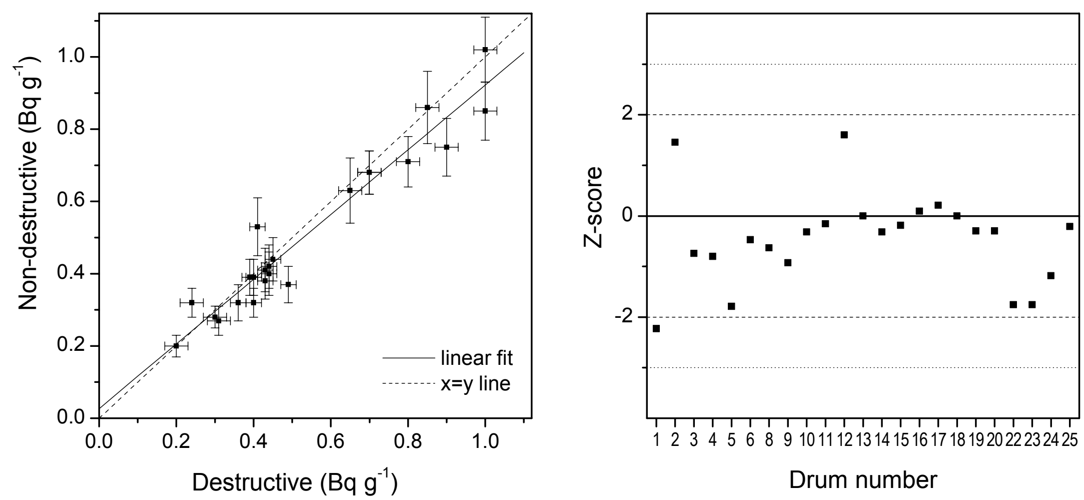

Fig. 3 ^{108m}*Ag activity concentration in the waste drums as estimated by the non-destructive drum assay and determined by sample analysis.*

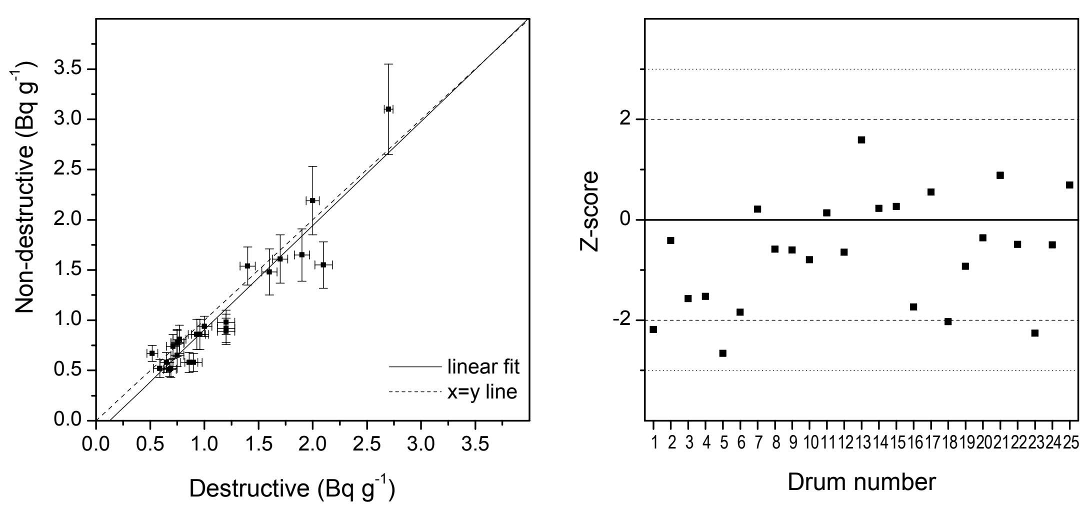

Fig. 4 ^{60}*Co activity concentration in the waste drums as estimated by the non-destructive drum assay and determined by sample analysis.*

the drum are well known, it has been shown that the non-uniformity of source, density of material and non-uniformity of matrix material may induce significant systematic errors in the determination of the activity. Since resin could be considered as a homogeneous matrix material, the main sources of bias in the interpretation of the analysis results could be identified as the non-uniform distribution of radioactivity within the drum, discrepancies in the filling height and differences in resin density. The magnitude of these errors was examined by Monte Carlo simulations of the drum and detector configuration. It was shown that for the worst case of activity inhomogeneity examined i.e. that of a line source at the center of the drum the induced error in the efficiency determination was of a factor of 1.9 and 1.6 at photon energies of 434 keV and 1332 keV, respectively.

Moreover, preliminary MCNP calculations showed the feasibility to examine source non-uniformity by measurement at different drum to detector positions.

The technique was verified by estimating radioactivity levels in 25 drums, containing ion exchange resin 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 discussed technique offers overall simplicity while minimizes labor and radiation protection requirements. Thus, it represents 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.

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