NON-DESTRUCTIVE TECHNIQUE TO VERIFY CLEARANCE OF PIPES

by

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A semi-empirical, non-destructive technique to evaluate the activity of gamma ray emitters in contaminated pipes is discussed. The technique is based on *in-situ* measurements by a portable NaI gamma ray spectrometer. The efficiency of the detector for the pipe and detector configuration was evaluated by Monte Carlo calculations performed using the MCNP code. Gamma ray detector full-energy peak efficiency was predicted assuming a homogeneous activity distribution over the internal surface of the pipe for 344 keV, 614 keV, 662 keV, and 1332 keV photons, representing Eu-152, Ag-118m, Cs-137, and Co-60 contamination, respectively. The effect of inhomogeneity on the accuracy of the technique was also examined. The model was validated against experimental measurements performed using a Cs-137 volume calibration source representing a contaminated pipe and good agreement was found between the calculated and experimental results. The technique represents a sensitive and cost-effective technology for calibrating portable gamma ray spectrometry systems and can be applied in a range of radiation protection and waste management applications.

Key words: research reactor, decommissioning, primary cooling system, clearance, MCNP code, key radionuclides, pipe

INTRODUCTION

The Greek Research Reactor (GRR-1) is a pool-type, light water moderated and cooled, heterogeneous reactor. GRR-1 went critical for the first time on June 1961. In 1971, the reactor was upgraded to 5 MW. A successful exploitation of the reactor was achieved through the development and utilization of experimental research facilities aiming to apply neutron techniques in research and technology and carry out interdisciplinary research in the areas of material science, condensed matter physics, applied nuclear physics and radioisotope production, health, environment, and cultural heritage studies. Available experimental facilities included neutron scattering techniques [1-3], irradiation rigs [4], neutron activation analysis [5], including capabilities for non-destructive analysis of large volume samples [6]. Moreover, throughout the operation of the reactor, expertise in nuclear technology and radiation protection was being further developed. The reactor was shut-down for refurbishment and modernization in 2004. The refurbishment of the reactor building was completed in 2008. In 2009, a program aiming to replace the primary cooling system (PCS) and to improve the design of the reactor and control system was initiated. Within this project, the decommissioning of the old PCS was one of the primary tasks. The said undertaking was part of preparatory activities for the decommissioning of the old GRR-1 primary cooling system composed of aluminum pipes, valves, two pumps, two heat exchangers, three delay tanks, a water purification system, and associated components.

The advanced prediction of radioactivity levels in contaminated components and equipment is of utmost importance when planning decommissioning activities, demonstrating compliance with clearance levels and acquiring appropriate approvals from the regulatory authorities. In the present work, a semi-empirical, non-destructive technique to evaluate the activity of γ-ray emitters (key radionuclides) in contaminated pipes is discussed. The technique was based on *in-situ* measurements by a portable NaI(Tl) γ-ray spectrometer. The efficiency of the detector for the complex pipe and detector configuration was evaluated by Monte Carlo calculations performed using the MCNP code. The γ-ray detector full energy peak efficiency was predicted assuming a homogeneous activity distribution over the

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internal surface of the pipe. However, the effect of activity inhomogeneity on the accuracy of the technique was also examined. The model was validated against experimental measurements performed using a Cs-137 volume calibration source prepared at our laboratory, representing the pipe detection geometry and a good agreement between calculated and experimental results established. The discussed technique represents a sensitive and cost-effective technology for calibrating portable γ-ray spectrometry systems. It can be applied in a range of radiation protection and waste management applications [7, 8].

EXPERIMENTAL

Model validation

A standard source representing an aluminum pipe with internal Cs-137 contamination was prepared. A filter paper was subdivided into 660 squares of 9 cm^2 each. In the center of each square, 0.05 ml (uncertainty in volume determination 10%) of Cs-137 acid solution (2 M HNO_3,) of (240 24) Bq/ml was dispensed, using a 0.5 ml pipette. The mean surface activity of the contaminated paper, eventually equaling the internal surface contamination of the standard pipe source, was 1.30 0.13 Bq/cm^2. The paper was positioned between two plastic sheets covering the internal surface of an aluminum pipe, 100 cm in length, with an external diameter of 20 cm and an internal diameter of 19 cm.

The measurements were performed using an Expluranium™ GR-130 mini SPEC portable γ-ray spectrometer equipped with a 38 mm in diameter and 57 mm in length, NaI(Tl) scintillation detector of a 7% resolution for the Cs-137 peak at 662 keV. The active center of the detector is considered to be at a depth of 4 cm from the detector surface. It was positioned at a distance of 25 cm and 35 cm from the geometrical centre of the standard pipe source and at right angles to the pipe's main axis of symmetry. Six measurements were performed at each distance, with the rotation of the pipe around its main axis. The duration of each measurement was 30 min. The spectral data accumulated were then transferred to a PC for analysis.

Detection limit

The detection limit (DL) in units of Bq/kg at each measurement configuration was evaluated by the following formula

$$DL \quad \frac{3\sqrt{B}}{eff_h \; A \, t \, M} \qquad (1)$$

where B are the counts of background at the spectrum area of the peak, for the measuring time, eff_h is the detector efficiency at the specific pipe-detector distance, A the abundance of γ-rays, t – the time of measurement [s], and M – the mass of pipe material [kg].

We wish to note that the detection limit determination at $3\sqrt{B}$ level was considered as adequate for the purpose of our study, *i. e*. radioactive waste management, and that it has been previously applied in other studies as well [9-11].

MONTE CARLO SIMULATIONS

A Monte Carlo simulation of the pipe and detector configuration was carried out using the MCNP5 code and cross-section data from the ENDF/B-VI library [12]. Both the code and cross-section library were obtained from the NEA Data Bank (France). Detector full energy peak efficiency was predicted.

The detector was modeled as a cylinder of sodium iodide of a 38 mm in diameter and 57 mm in length. Our model geometry included a homogeneously distributed surface source on the internal surface of an aluminum pipe. Pipe dimensions were 100 cm in length, 19.8 cm in internal diameter, and 21.6 cm in external diameter. Runs were performed for 344 keV, 614 keV, 662 keV, and 1332 keV photons, representing the main γ-ray lines of the target nuclides Eu-152, Ag-108m, Cs-137, and Co-60, respectively. 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 range equal to the peak integration area. A relative error of less than 3% was achieved in all predicted efficiencies.

Simulations were also performed to examine the effect of inhomogeneous activity distribution within the pipe. Two activity distributions were modelled; representing the worst envisaged cases of inhomogeneous activity distributions. The results obtained were compared with those from the homogeneous activity distribution. The examined cases represented activity distributed over a ring of a width of 1 mm on the internal surface of the pipe, at the top and at the middle of the pipe, respectively (fig. 1). The NaI crystal active cen-

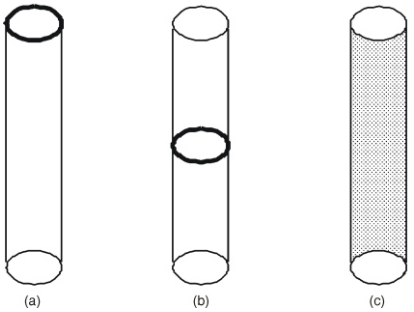

Figure 1. Modeled activity distributions
(a) ring at the top of the pipe, (b) ring at the middle, (c) homogeneous distribution over the inner surface

ter was positioned at 15.5, 25.5, 30.5, 35.5, 40.5, and 45.5 cm from the geometrical center of the pipe, at the mid-height level of the pipe, with its axis at right angles to the pipes' main axis of symmetry. Detector efficiency for the gamma lines of interest was predicted as a function of the distance of the detector from the geometrical center of the pipe.

RESULTS

Model validation

Table 1 shows the mean value and the standard deviation obtained from six measurements (with the rotation of the pipe around its main axis) for two pipe-detector distances of the standard volume source. We note that the standard deviation of the six measurements at each position is comparable to the statistical error of the measurement at each distance. The calculated activities were in agreement with the nominal activity of the standard source. The calculated to nominal activity ratio was 0.91 0.10 and 0.93 0.11 for 662 keV photons at 25 and 35 cm pipe-to- detector distance, respectively.

Table 1. Surface specific activity for Cs-137 [Bq/cm^2]

Nominal	Evaluated	
	Pipe-detector distance [cm]	
	25	35
1.30 0.13	1.18 0.05	1.21 0.08

Simulations

Figures 2 to 5 represent the predicted MCNP detector full energy peak efficiencies for 344 keV, 614 keV, 662 keV, and 1332 keV photons, respectively, for activity distributions shown in fig. 1. In cited figures, the predicted detection limit for each modeled configuration is also shown. It can be observed that for 60 min counting time and a measurement distance of 30 cm, a minimum detection limit of 0.55 Bq/g for Eu-152, 0.02 Bq/g for Ag-108m, 0.02 Bq/g for Cs-137, and 0.015 Bq/g for Co-60 was obtained. Figure 6 shows the ratio of predicted full energy peak efficiencies of the inhomogeneous distributions to the homogeneous distribution for 662 keV photons (Cs-137) as a function of source-to-detector distance. It was shown that at the distance of 30 cm for a ring distribution of activity at the middle of the pipe, fig. 1(b), and for ring activity distribution at the top of the pipe, fig. 1(a), the ratio was 1.5 and 0.4, respectively. Therefore, an over-estimation of about 50% and an under-estimation of about 60% will be encountered in the special cases of activity distribution with a ring in the middle or at the top of the pipe, respectively. Similar results were obtained for other radionuclides examined in this study.

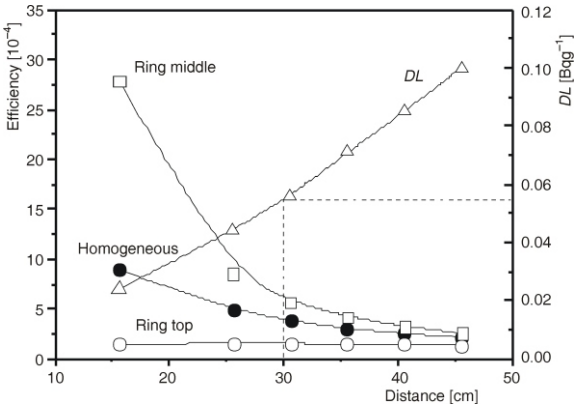

Figure 2. Predicted detector peak efficiency for homogeneous and inhomogeneous distribution of Eu-152 (344 keV) as a function of source-to-detector distance and evaluated measurement detection limit for 60 min counting time

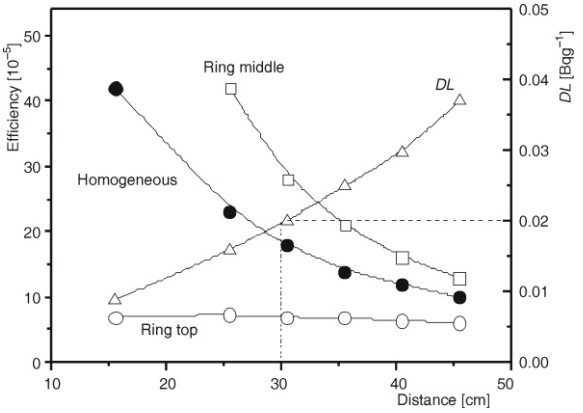

Figure 3. Predicted detector peak efficiency for homogeneous and inhomogeneous distribution of Ag-118m (614 keV) as a function of source-to-detector distance and evaluated measurement detection limit for 60 min counting time

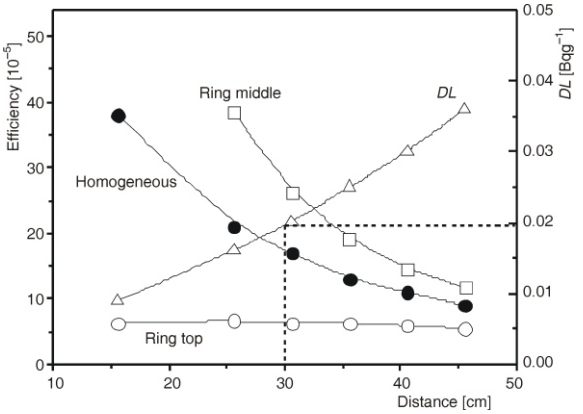

Figure 4. Predicted detector peak efficiency for homogeneous and inhomogeneous distribution of Cs-137 (662 keV) as a function of source-to-detector distance and evaluated measurement detection limit for 60 min counting time

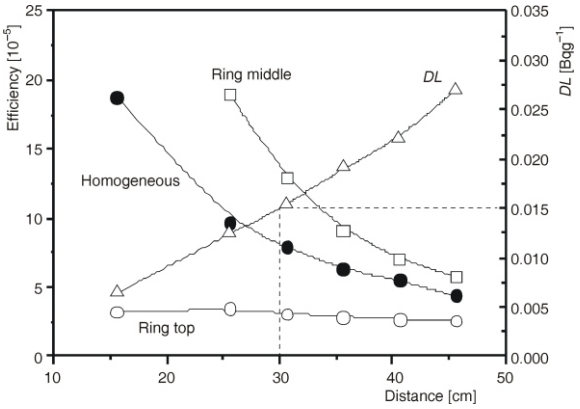

Figure 5. Predicted detector peak efficiency for homogeneous and inhomogeneous distribution of Co-60 (1332 keV) as a function of source-to-detector distance and evaluated measurement detection limit for 60 min counting time

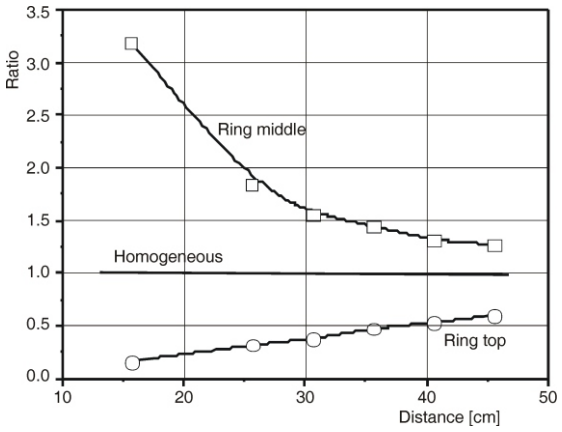

Figure 6. Ratio of inhomogeneous to homogeneous activity distributions peak efficiency as a function of source-to-detector distance for Cs-137

The mass specific general clearance level for Eu-152, Ag-108m, and Co-60 is 0.1 Bq/g and for Cs-137 is 1 Bq/g [13]. Therefore, the predicted detection limits of the measurement were found to be lower than the clearance level by a factor of 50 for Cs-137, 5 for Ag-108m, and 7 for Co-60. For Cs-137, Co-60, and Ag-108m, these detection limits were adequate for their determination, based on a 30 cm working distance, with the biases being acceptable, as well. However, for Eu-152, the detection limit must be further reduced by increasing, for example, the time of measurement.

GENERAL DISCUSSION AND CONCLUSIONS

A semi-empirical, non-destructive technique combining gamma spectrometry and Monte Carlo simulations using the MCNP code for the determination of key radionuclides in pipes was discussed here. The technique was validated against experimental measurements performed using a standard source representing a pipe internally contaminated with Cs-137. The results showed satisfactory agreement within 1σ. The effect of activity inhomogeneity on the accuracy of the technique was examined as a function of the pipe-to-detector distance. The measurement distance between pipe and detector was chosen so as to allow for an adequate detection limit for key radionuclides, while taking into account the clearance criterion and the measurement bias due to possible inhomogeneity in the distribution of the activity. These biases can be reduced by, for example, reducing the length of the pipes.

The chosen measurement distance for a pipe of a 19.8 cm inner diameter, 0.9 cm wall thickness and 100 cm length, internally contaminated with Ag-108m, for a 60 min counting time, was 30 cm. At this distance, a detection limit of 0.02 Bq/g was achieved. The detection limits for Eu-152, Cs-137, and Co-60, for the same counting time and measurement distance, were 0.55, 0.02, and 0.015 Bq/g, respectively. It was estimated that, in worst cases of source activity inhomogeneities: (a) for a ring source at the top of the pipe, the under estimation is 60% and (b) for that of the ring source at the middle of the pipe, the over estimation is 50%.

The discussed technique was shown to be sensitive enough for the determination of key radionuclides. Furthermore, it is a cost-effective method for calibrating γ-ray monitoring systems that can be applied in a range of radiation protection and waste management applications. In particular, it is to be applied for the predictions concerning internal surface pipe contamination and the verification of clearance levels of the Greek research reactor primary cooling system pipes, scheduled to be decommissioned for replacement and renovation. Key radionuclides will be evaluated and appropriate scaling factors will be used for the confirmation of clearance criterion or for waste material characterization [14].

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НЕДЕСТРУКТИВНА ТЕХНИКА ЗА ПРОВЕРУ РАДИЈАЦИОНЕ ЧИСТОЋЕ ЦЕВИ

Размотрена је једна семиемпиријска недеструктивна техника за процену активности извора гама зрачења у контаминираним цевима. Техника је заснована на *in situ* мерењима портабл NaI гама спектрометром. Ефикасност детектора за цеви и детекторска конфигурација процењени су Монте Карло рачунима обављеним коришћењем MCNP програма. Ефикасност гама детектора при укупном енергетском пику била је процењена претпостављајући хомогену расподелу активности по унутрашњој површини цеви за фотоне енергија од 344 keV, 614 keV, 662 keV и 1332 keV, који репрезентују Eu-152, Ag-118 m, Cs-137 и Co-60 контаминацију, респективно. Испитан је, такође, утицај нехомогености на тачност мерне технике. Модел је потврђен експерименталним мерењима обављеним коришћењем Cs-137 запреминског калибрационог извора који представља контаминирану цев, и нађена је добра сагласност израчунатих и мерених резултата. Ова техника представља осетљиву технологију прихватљиве цене за калибрацију преносивих гама спектрометарских система која може да се примени у заштити од зрачења и управљању отпадом.

Кључне речи: истраживачки реактор, декомисија, примарни систем хлађења, радијациона чистоћа, MCNP програм, кључни радионуклиди, цев