Characterization of Spent Filters from the Water Polishing System of IEA-R1 Reactor – 15451

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ABSTRACT

The aim of this paper is to report studies performed to determine the activity of gamma emitters present in polypropylene cartridge filters from the polishing system of the IEA-R1 Reactor cooling-water primary circuit. The activities were calculated using the dose rates measured with handheld detectors, after the ratios of the emission rates of photons were evaluated by gamma spectrometry, and the Point Kernel method, which correlates the activity of a source with the dose rates at various distances. The method described can be used to determine routinely the radioactive inventory of these filters, avoiding the necessity of destructive radiochemical analysis, or the necessity of calibrating the geometry of measurement.

INTRODUCTION

Expanded polypropylene filter cartridges are used to retain particulate matter suspended in the water of the primary cooling circuit of the IEA-R1 reactor. These filters are part of the cooling water polishing system, which also includes beds of ion exchange resin and activated charcoal.

The IEA-R1 reactor is a pool-type research reactor, operating between 2 and 5 MW that uses water as coolant, moderator and biological shield. Besides research, it is used for production of radioisotopes and irradiation of samples with neutron and gamma beams. It is located in the Nuclear and Energy Research Institute at the University of Sao Paulo campus.

When filters become saturated and are unable to maintain the flow within the established limits, they are replaced and disposed of as radioactive waste. After a few weeks of decay, they are sent to the Radioactive Waste Management Facility (GRR), which is responsible for the treatment and temporary storage of radioactive wastes generated by IPEN as well as the wastes generated by several other institutions that use radioisotopes in Brazil.

One of the first steps of the waste management is the primary characterization of the waste, which is necessary to guide treatment processes and to establish the level of the radiation protection required. The main objective of the primary characterization is to determine the isotopic inventory of the wastes [2].

The final characterization, in turn, is intended to meet the safety objectives set by the regulator for transportation and acceptance of the waste for disposal in a repository [3]. Safety analysis of repositories require that the radionuclide inventory of disposed materials is known in order to ensure the radiation safety objectives in the long term are achieved [4].

The filter cartridges used in the IEA-R1 reactor are made of expanded polypropylene, with porosity equivalent to 10μm; they have a cylindrical-annular geometry, 65 mm outer diameter, 25 mm inner diameter, 508 mm length and the spent, dry filters weight about 350 g.

These filters are collected and transported in 200 L drums and are stored as radioactive waste in the waste storage of GRR.

The radionuclides present in the reactor cooling system that are retained in the filters are the activation products, fission products and transuranic elements contained in particulate material suspended in the cooling water.

The development of a primary characterization method, for determining the radioactive inventory of these filters is required for routine operation in the GRR. A protocol for the primary characterization of the material, so that the radionuclides present and their respective activities can be known in a simple and accurate manner, is a contribution to the GRR operational routine. One alternative method is the radiochemical analysis of slices taken from each filter, what presents the disadvantage higher exposures of personnel and contamination risks. Another alternative method is the calibration of the measurement geometry of a gamma spectrometer, which requires the production of a 'standard filter'.

This work aims at presenting the method developed to estimate the activity of gamma emitters in filter cartridges, using the method of Point Kernel and the dose rates measured in filters with handheld gamma detectors. The ratio of emission rates between the detected gamma photons must be known and can be determined by gamma spectrometry, but the method avoids the necessity of calibrating the detectors for the measurement geometry.

METHODS

From a group of several dozen filters already collected as waste, a sample of 15 filters was selected with dose rates in contact about 0.3 mGy/h or less, using a survey meter (Automess GmbH, model 6150 AD). This value was chosen to keep low the doses in the individuals during the subsequent measurements and, at the same time, allowing the determinations with reasonable accuracy, for a sample of this size. Each selected filtration unit was individually wrapped in polyethylene bags and labeled.

The homogeneity of each filter was checked to guarantee that it is possible to apply the calculation method, which assumes that the distribution of activity is uniform. To this purpose, a shielding was constructed with lead bricks, in the form of a tunnel, so that each filter could be completely shielded in the horizontal position. The shielding was long enough to allow the filter to move along its axis without loss of shielding. A beam-hole 1 cm in diameter in the central brick allowed the radiation detector to measure the radiation emitted by a thin slice of the filter each turn. The readings were taken at every 3 cm, totaling seventeen slices for each filter. According to IAEA [6], a waste can be considered homogeneous if the concentration of one radionuclide used as indicator, in different parts of the waste, is within the range of ± 30% around the average concentration.

After this, the dose rates were measured at the distances of 20 cm, 40 cm and 60 cm from the surface of the filter in the median plane of the cylinder, using the Radiagem 2000 (Canberra, Radiagem 2000 Personal Portable Dose Rate and Survey Meter), and the 6150 AD Automess detectors. The two detectors were used as a means of comparing results and detecting any discrepant values.

The filters were then positioned near a hyperpure germanium detector (Ortec, HPGe, model EPCG-15-190-R), calibrated in energy. Each filter was measured during 600 seconds. The photopeak heights were corrected by the detector energy efficiency curve and the ratios between the peak heights were used as an estimate of the ratios of emission rates between any pair of photon energies of radionuclides in the filter. The ratios were input data for calculating the dose rates expected in the measurement positions, assuming

that the proportions between the emission rates of photons of each energy are the same proportions that were detected. This simplification is acceptable since the differences in the self-absorption of radiation in the filters and the absorption in the air between the filter and the detector can be neglected, for the energies considered.

Using the method of Point Kernel described by Rockwell [5] or the MicroShield® software and the proportions of the emitted intensities of each photon energy identified in the gamma spectrometry, the expected dose rates were calculated at each of the measurement distances above. In these calculations, the MicroShield is used in the mode where the individual photon energies and the emission rates are specified.

Each photon contributes to the fluence at the measuring position, so that the dose rate at a point P (See Figure 1) is given by:

$$\dot{D} = \sum_i \Phi_i \cdot G_i$$

Where:

$\dot{D}$: dose rate at the point P, given in $Gy.h^{-1}$,

Φ_i: flow of photons with energy i at the P position, given in $cm^{-2}.s^{-1}$

G_i: dose factor per unit of flow of photons with energy i, given in $Gy.\ h^{-1}.cm^2.s$

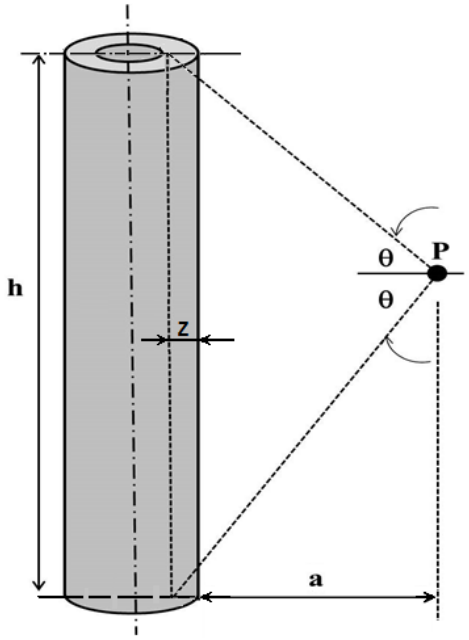

Figure 1: Geometry of the source and the detector for dose measurements at point P

The flow of photons of each energy, expected at the measuring position, is calculated by:

$$\Phi_i = \frac{B_i \cdot S_{Vi} \cdot R_o^2}{2(a + Z_i)} \cdot F_i(\theta, b)$$

Where:

B is the build up factor, calculated for each energy i,

Sv_i is the emission rate of photons of energy i, per unit of volume, given in $s^{-1}.cm^{-3}$

R_0 is the radius of the filter, given in cm;

a is the distance from the measuring position to the filter surface, given in cm;

Z_i is the self-attenuation distance of each photon of energy i inside the filter, given in cm;

$F_i(\theta, b_i)$ is the integral

$$F_i(\theta, b_i) = \int_0^{artg(\frac{h}{2a})} e^{b_i \, sec\theta} d\theta$$

in which $b_i = \mu_{Si} Z_i$

where μ_{Si} is the linear attenuation coefficient for the photons of energy i in the filter, in cm^{-1}.

The calculation of the dose rate at the measuring positions can be done with any values of Sv_i, as long as the proportions between the emission rates of any pair of photon energies agree with the emissions rates determined previously.

Finally, the Sv_i values are adjusted so that the calculated dose rate coincides with the measured value. The adjustment is done by averaging the proportions between the values measured at three distances from the filter and the values that were calculated with the initial estimates of the Sv_i values.

To check the accuracy of the method, the new calculated values of Sv_i of each energy are translated into the activity of the corresponding radionuclides, considering the yield on the photons in the decay, and the activities are used to calculate the dose rate at the measurement positions, using Microshield. In these calculations, the software is used in the mode where the radionuclides and the activities are specified.

RESULTS

Some results of the homogeneity tests are shown in the graphs of Figure 2 for two filters of the sample, identified as A and B, as an example. In 4 filters sampled, the results were within ± 9,5% interval around the average values of the count rate obtained.

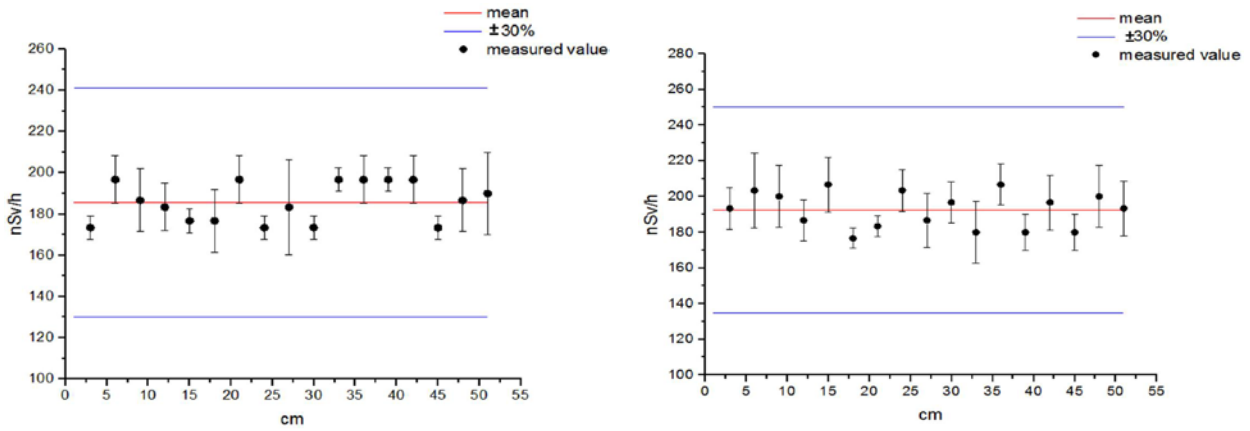

Figure 2: Results of the homogeneity test in two filters of the sample.

The gamma spectrometry identified the same gamma emitters in all the 15 filter cartridges assessed. The identified radionuclides are shown in Table 1 with their respective sources in the reactor core.

Table 1: Radionuclides present in the filters

Radionuclide	$T_{1/2}$	Eγ (keV)	Iγ (%)	Formation Reaction	Origin
^{60}Co	5,27 a	1173	99,9	$^{60}Ni(n,p)^{60}Co$	Stainless Steel AISI 304 and Cr-Ni in the lining of the reactor pool.
		1333	100	$^{61}Ni(n,np)^{60}Co$	
				$^{63}Cu(n,a)^{60}Co$	Alloys ASTM 1060, 6262 and 6061 from the core structures.
^{108m}Ag	418 a	723	91,3	$^{107}Ag(n,g)^{108m}Ag$	Neutron absorbing alloy of the control rods.
		614	91,2	$^{109}Ag(\gamma,n)^{108m}Ag$	
		434	90,7	$^{109}Ag(n,2n)^{108m}Ag$	
^{110m}Ag	249,9 d	658	94,7	$^{109}Ag(n,g)^{110m}Ag$	Neutron absorbing alloy of the control rods.
		885	72,9	$^{113}In(n,a)^{110m}Ag$	
		938	34,3	$^{110}Cd(n,p)^{110m}Ag$	
		1384	24,3	$^{111}Cd(n,np)^{110m}Ag$	

The ^{137}Cs was not observed in any of the filters, although it is one of the main radionuclides found in the reactor wastes. This indicates that the cesium is fully dissolved in the water and is not detectable in any precipitate, nor is adsorbed on the suspended particulate material.

The measured dose rates and the calculated dose rates using the initial estimate of Sv_i, are shown in the graphs of Figure 3 for two filters of the sample, as an example of the results.

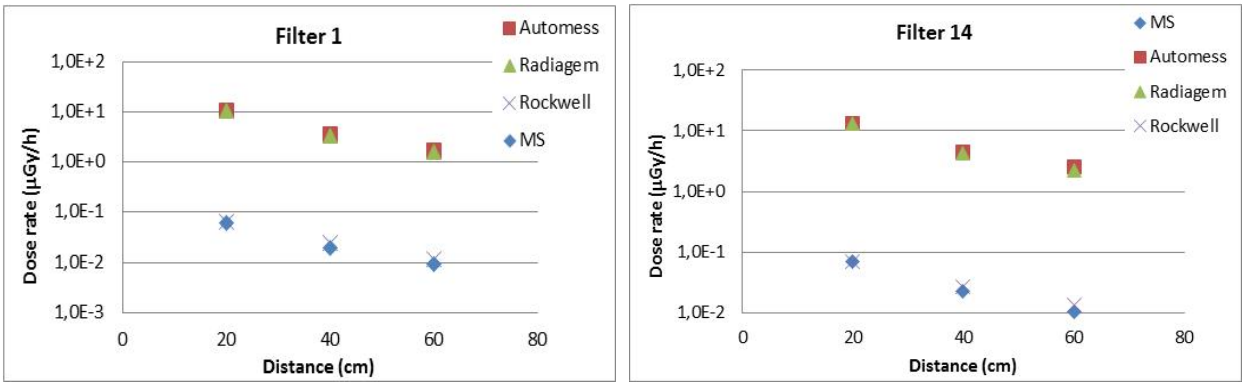

Figure 3: Measured dose rates and the calculated dose rates using the initial S_{vi} estimate.

The average ratio between the measured and the calculated dose rates at the three distances from the surface for each filter was used to refine the initial estimates of S_{Vi}. The new values were used to calculate the activities of the corresponding radionuclides and this value is taken as the end point of the method. Using these activities in the MicroShield in the mode 'radionuclide & activity', the expected dose rates were calculated and compared with the measured values. The results shown in Figure 5 point to an internally consistent method. However, the uncertainties of the method were not yet determined. The main contribution to the end-point calculation errors is the accuracy of the survey meters that is 20% of the measured dose rates, and the different attenuations of the gamma photons in the filter and in the air for different energies, which were disregarded in the calculations.

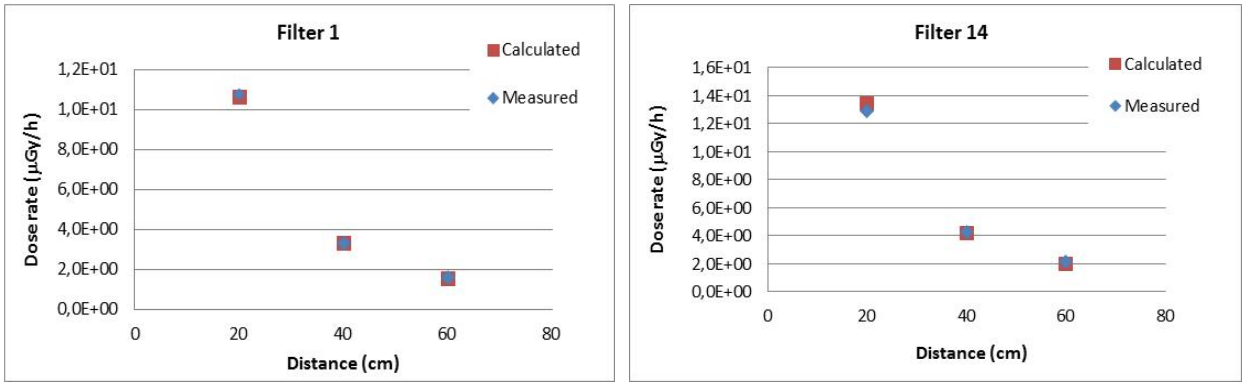

Figure 4: Measured and calculated dose rates for filters 1 and 14 of the sample, as an example of the method's internal consistency.

FINAL REMARKS

The measured dose rates at three distances from the filters and the ratios of the emission intensities of each photon energy identified by gamma spectrometry were used to calculate the activities of the corresponding radionuclides present in filter cartridges from the IEA-R1 Research Reactor cooling-water polishing system.

Using a gamma spectrometer calibrated in energy, a handheld survey meter, and a computational software it was possible to determine the activities of a sample of 15 spent filters stored as radioactive waste at the Radioactive Waste Management Department of IPEN. The accuracy of the method is yet to be determined

and will be estimated by comparing the present results with the results of a Monte Carlo detector calibration method and with the results of a radiochemical analysis of sliced samples that are under development.
The method described herein can be used to determine routinely the radioactive inventory of these filters, avoiding the necessity of destructive testing and radiochemical analysis, or the necessity of calibrating the measurement geometry of the detectors.

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