Radiochemical Method for Characterization of the Filter Cartridges from the IEA-R1 Reactor – 16324

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ABSTRACT
The filter cartridges used in water purification system of the IEA-R1 research reactor become radioactive waste after the end of their useful life. The characterization of this waste is one step in their management, which aims at identifying and quantifying the radionuclides present, including those known as "difficult to measure" (DTM) radionuclides. Wastes from nuclear reactors contain fission and activation products and transuranic elements, only few of them emitting gamma radiation measurable by simple gamma spectrometry methods. In routine waste management activities, the concentrations of the DTM can only be estimated by indirect methods such as scaling factors. The method of the scaling factors uses empirically determined proportions between the concentrations of DTM radionuclides and that of easily measurable gamma emitters called key nuclides (KN), to calculate DTM concentrations. Determining the scaling factors for the filter cartridges is a hard task when the number of samples is large, not only because the preparation of samples is difficult but also because the number of radionuclides to analyze is large. If it can be demonstrated that DTM and KN are both distributed evenly in the filters, less samples from each filter are necessary, reducing lab work. The aim of this paper is to present the results of the assessment of homogeneity along the filter cartridges by gamma spectrometry of chemically reduced samples. Five thin slices of five filters were burnt and the ashes quantitatively dissolved and the activity concentration of the solutions determined by gamma spectrometry.

INTRODUCTION
The Nuclear and Energy Research Institute (IPEN–CNEN/SP) located at the University of São Paulo, Brazil, operates the IEA-R1 reactor since 1957. It is an open pool-type nuclear research reactor, operating at 2 to 5 MW that uses water as coolant, moderator and biological shield. Research, production of radioisotopes and irradiation of samples with neutron and gamma beams are the main activities developed at the facility.

Polypropylene filter cartridges are part of the water treatment system of the reactor and become radioactive as they remove suspended solid material from the cooling water during the operation of the reactor. Quality control of the water indicates the necessity of periodic replacement of these filters. After some weeks awaiting decay of very short-lived isotopes, they are transported to the Radioactive Waste Management Facility (RWMF) at IPEN, for storage and treatment. The RWMF is responsible for treatment and interim storage of the radioactive wastes generated by IPEN or received from many radioisotope users in the country.

Formerly, the filters were drummed and compacted at the RWMF without any concerns about the inventory. However, present regulations require that the radioactive content of each drum be determined and stated.

There are a number of techniques that can be used to obtain the radioisotope content but to choose the most suitable method to use, it is necessary to take into consideration factors such as the type of radiation emitted, the costs and availability of measuring instruments, the dose rates involved in taking and handling waste samples, the physical state and homogeneity of the waste, among others (IAEA, 2007).

Annually, the replacement of the filters from the reactor's water treatment system generates about 36 units with initial contact dose rates in the range of tens of millisievert per hour. Therefore, the development of a method for the radioisotopic characterization of these filters is required for routine operation in the RWMF, to determining the radioactive inventory of waste packages.

Wastes from nuclear reactors contain fission and activation products and transuranic elements, only few of them emitting gamma radiation measurable by simple gamma spectrometry methods.

The radionuclides whose concentrations cannot be measured by direct gamma spectrometry are called Difficult To Measure (DTM).

In routine waste management activities, the concentrations of the DTM can only be estimated by indirect methods, such as scaling factors. The method of the scaling factors uses empirically determined proportions between the concentrations of DTM radionuclides and that of easily measurable gamma emitters called key nuclides (KN), to calculate DTM concentrations.

Determining the scaling factors for the filter cartridges is a hard task because the number of samples is large, the preparation of samples is difficult and the number of radionuclides to analyze is large. If it can be demonstrated that DTM and KN are both distributed evenly in the filters, less samples from each filter are necessary, reducing lab work.

The aim of this paper is to present the results of the assessment of homogeneity along the filter cartridges by means of gamma spectrometry of chemically reduced samples.

The activity concentration was determined with samples solutions obtained after destruction of the filters. Besides gamma analysis, a radiochemical analysis methodology is under development to identify and quantify alpha and beta emitters. The final objective is to establish the Scaling Factors or the Correlation Functions for this kind of waste (Taddei *et al*., 2015).

METHODS

Five thin slices of five randomly selected filters from drums with uncompacted wastes were burnt and the ashes quantitatively dissolved and the activity concentrations of the solutions were determined by gamma spectrometry.

The activity concentration was measured in the solutions obtained after dissolution of the samples.

Representative samples were taken, according to the scheme shown in Fig. 1. Five, 2 cm thick slices were cut from evenly distributed parts of each filter,

and analyzed to assess the homogeneity of the filters. The activity of the slices were not directly measured by gamma spectrometry due to the difficulty in preparing a calibration standard.

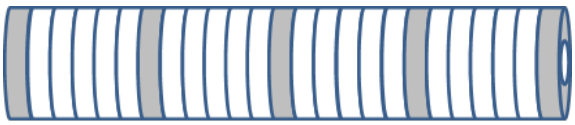

Figure 1. Scheme adopted in the sample collection: filters were marked to define 17 slices and those identified by a darker color were cut and taken as samples

Each filter slice taken as sample was cut in half and one part were weighed and calcinated separetly in oven for 24 hours at 400 °C. The ashes were dissolved in a teflon beaker with 3 aliquots of 20 mL of acqua regia, on a heater plate at 250 °C, each portion being added after the dryness of the sample. Then, 3 aliquots of 10 mL of 69-72% $HClO_4$ and 10 mL of 65% HNO_3 were added and the solution was heated until dryness. Following the same procedure, 3 aliquots of 5 mL HF 48% and 10 mL of HNO_3 65% were added. Residues of acqua regia, HF and $HClO_4$ were eliminated with 3 aliquots of 5 mL of HNO_3, H_2O_2 and 2 mL of deionized water.

The samples were then completely dried, cooled and the salts were dissolved in about 20 mL of 8M HNO_3 solution, transferred to 100 mL volumetric flask and the volume then completed with the same solution.

The activity concentrations were determined by gamma spectrometry of 10 mL aliquots. A Gamma Spectrometry System consisting of a HPGe (High Purity Germanium) detector, Model GX 2518 from Canberra Industries was employed.

DISCUSSION

The mean values of the total activity of the filter cartridges obtained by gamma analysis of samples are shown in Tab. I. The analyses revealed the presence of the radionuclides ^{60}Co, ^{108m}Ag and ^{110m}Ag.

TABLE I. Mean values of the total activity content of filter cartridges

Filter id	Activity (Bq)		
	Co-60	Ag-108m	Ag-110m
6	7.7E+05	2.1E+05	5.8E+04
9	8.3E+05	2.3E+05	6.4E+04
10	8.1E+05	1.6E+05	7.4E+04
12	8.3E+05	1.8E+05	7.3E+04
15	7.5E+05	2.1E+05	2.5E+05

To assess the homogeneity of the filters, the activity concentrations determined in each filter slice were converted into the corresponding Z-scores, by dividing the differences between each result and mean value of one filter, by the standard deviation of the results of the five slices. The results are shown in Fig. 2.

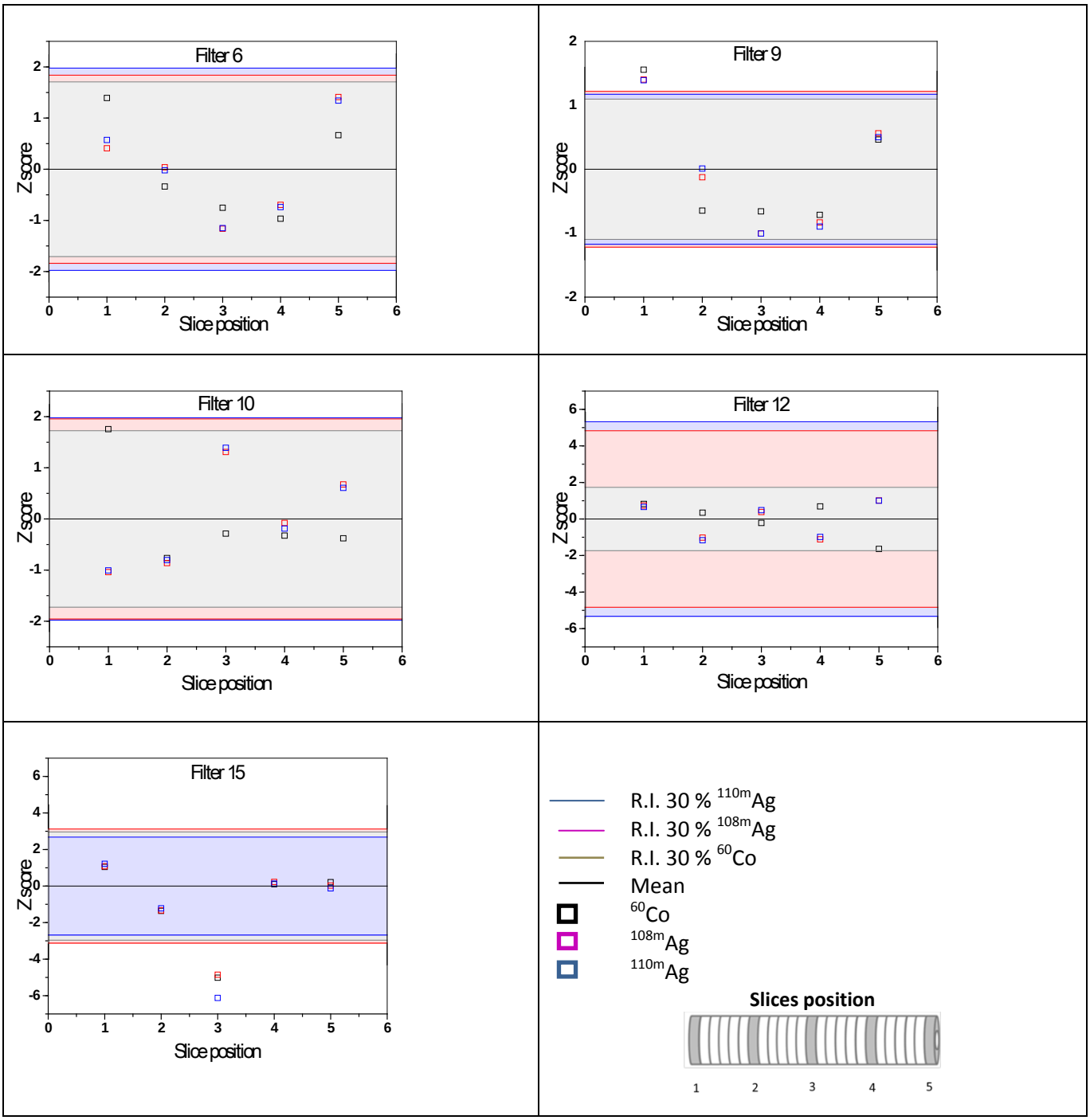

Figure 2. Evaluation of the homogeneity by gamma spectrometry

It was considered for the evaluation of homogeneity that for stable waste streams, measuring one or more key nuclides may be sufficient to check the homogeneity 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 (IAEA, 2007). In this way, despite of some fluctuation observed, the method showed results that are within this relative interval, indicating homogeneity in each filter cartridge.

CONCLUSIONS

This paper describes preliminary results of the characterization of filter cartridges released as radioactive waste from the IEA-R1 research reactor located at IPEN-CNEN/SP.

This study identified the key gamma emitting radionuclides and revealed a homogeneous distribution of the activity along the filter cartridges. This result is important to define the sample size for the determination of the difficult to measure radionuclide in the forthcoming steps in the characterization program. One slice per filter can be considered as a representative sample of the waste, implying in a smaller number of samples to be analyzed.

The next step is to measure the concentrations of the DTM and determine the scaling factors or correlation functions, by combining the results presented in this paper with those that will be obtained from radiochemical analysis of the samples.

However, before the analyses, a careful planning of the sampling will be carried out in order to define the ideal number of filter to be measured, taking into consideration the occupational doses that will be incurred by facility operators and laboratory personnel.

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