2015 International Nuclear Atlantic Conference - INAC 2015
São Paulo, SP, Brazil, October 4-9, 2015
ASSOCIAÇÃO BRASILEIRA DE ENERGIA NUCLEAR - ABEN
ISBN: 978-85-99141-06-9

CHARACTERIZATION OF FILTER CARTRIDGES FROM THE IEA-R1 REACTOR BY RADIOCHEMICAL METHOD

Bianca Geraldo[1], Roberto Vicente[1], Robson J. Ferreira[1], Marcos M. Goes[1] and Júlio T. Marumo[1]

[1] Instituto de Pesquisas Energéticas e Nucleares (IPEN / CNEN - SP)
Av. Professor Lineu Prestes 2242
CEP 05508-000 São Paulo, SP
bgeraldo@ipen.br

ABSTRACT

The filter cartridges used in water purification system of research nuclear reactor IEA-R1 are considered radioactive wastes after their useful life. The characterization of these wastes is one of the stages of management, which aims to identify and quantify the radionuclides present, including those known as "difficult to measure" (DTM) radionuclides. Establish a radiochemical analysis methodology for this type of waste is a difficult job, not only by the application of these techniques, but also by the amount of radionuclides that should be analyzed. In the waste produced in a nuclear reactor, the most important radionuclides are fission products, activation products and transuranic elements. Since these radionuclides emit gamma radiation not measurable in its decay process and consequently are difficult to measure, their concentrations can be estimated by indirect methods such as scale factors. This method is used to evaluate the DTM concentration, which is represented by alpha and beta nuclides using the correlation between them and the radionuclide key, a gamma emitter. The objective of this work is to describe a radiochemical analysis methodology for gamma emitter nuclides, present in the filter cartridges, evaluating the activity and concentrations by destructive assays. At the same time, two studies have been performed by non-destructive assays, the first one based on dose rates and the point kernel method to correlate the results and the second one based on calibration efficiency with Monte Carlo method. These studies belong to the radioactive waste characterization program that has been conducted at the Waste Management Laboratory of Nuclear and Energy Research Institute, IPEN-CNEN/SP.

1. INTRODUCTION

The Nuclear and Energy Research Institute (IPEN-CNEN/SP) located at the University of São Paulo, Brazil, operates a pool-type nuclear research reactor at 2 to 5 MW since 1957. Some radioactive wastes arising in the routine operation of the facility are stored on site and still require characterization and treatment. One of them is polypropylene filter cartridges from the water cooling system. These filters become radioactive by removing suspended solid material from the cooling water when the reactor is in operation. The control of water quality indicates the necessity of periodic replacement of these filters, which are transferred to the IPEN-CNEN/SP Waste Management Department after a cooling period [1].

The waste management involves various activities and one important step in the management of this waste is the primary characterization, in which its physical and chemical properties are determined, the radionuclides present are identified and their activities are measured [2].

There are a number of techniques that can be used to obtain the radioisotope content of the waste, such as radiochemical analysis of waste samples, gamma scanning of the whole waste package or gamma spectrometry of each filter cartridge, mathematical modeling of the waste generating process, etc. To choose the must 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 [3].

To establish a radiochemical analysis methodology for this type of waste is a hard job, not only for the difficult chemical reduction of the filter material and the radiochemical separation of the radionuclides, but also by the number of radionuclides that must be determined. However, it is the more comprehensive and accurate.

Over twenty radioisotopes of fifteen different elements are radiologically relevant and must be determined in the waste to comply with disposal acceptance criteria.

The most important radionuclides present in this radioactive waste are the activation products: ^{55}Fe, ^{60}Co, ^{59}Ni and ^{63}Mi, ^{108m}Ag and ^{110m}Ag; the fission products: ^{3}H, ^{14}C, ^{90}Sr, ^{99}Tc, ^{129}I, ^{135}Cs and ^{137}Cs; and the actinoids: ^{234}U, ^{235}U, ^{238}U, ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{241}Am, ^{242}Cm.

Several in the list, are alpha and beta emitter that can only be analyzed by the radiochemical method. These radionuclides are called difficult to measure radionuclides (DTM). The gamma emitters that can be determined by gamma spectrometry of samples and gamma scanning of the whole waste volume are called key radionuclides (KN). In the above list, the KN present in the filter cartridge are ^{60}Co, ^{108m}Ag and ^{110m}Ag [4, 5].

To overcome the problem of the high costs and the technical difficulties in the routine characterization of the DTM, the method of scaling factors is being tried. The method of scaling factors (SF) is a technique in which the concentrations of the DTM are measured in a few waste batches by radiochemical methods and then divided by the measured concentrations of the KN. The ratios ate the scaling factors. Then, the content of DTM in subsequent waste batches is estimated by multiplying the SF by the measured concentrations of the KN in the new batches. This method has good applicability for waste streams that are generated continuously without great variations, i.e. homogeneous wastes [6, 7].

The objective of this paper is to describe a radiochemical analysis methodology for the determination of gamma emitting radionuclides, present in the filter cartridges, evaluating the activity concentrations by destructive assays.

At the same time, two studies were conducted using non-destructive testing, at first, the activities of gamma emitting radionuclides present in the filters was determined using gamma spectrometry, dose rate and the Point Kernel method for correlating the measurement results since the second study presents a technique using Monte Carlo method associated with gamma spectrometry, based on the calibration efficiency obtained through the Monte Carlo method. These studies are part of a radioactive waste characterization program being conducted by IPEN-CNEN / SP [8, 9].

In this work, we were selected for study the filter cartridges, which in turn have a replacement more often, and in each exchange are provided six cartridges. Annually are generated 36 units, a lot of this material is stored on the Management of Radioactive Waste storage (MWR) at IPEN, waiting for the characterization of radionuclides.

2. MATERIALS AND METHODS

2.1. Sample Collection

The filter cartridges from the IEA-R1 research reactor water treatment system are packed in various drums with 200 L of capacity, previously stored in polyethylene bags. These drums are stored in the Waste Management Radioactive (WMR) at IPEN, located on the campus of the University of São Paulo.
To obtain representative samples, it was determined that would be cut from each filter, 5 slices each 2 cm, evenly distributed, so that thereby the homogeneity of the filters is measured, as shown in Figure 1.

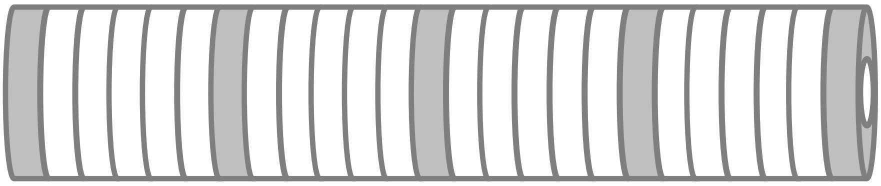

Figure 1: Scheme adopted in the collection of filter cartridges

2.2. Sample Preparation

After sampling, each filter slice was cut in half, then the samples were weighed and kept in oven for 24 hours at 400 °C to eliminate the organic matter. For the dissolution of the sample was added into a teflon beaker of 250 mL total sample mass, 3 portions of 20 mL of $HCl:HNO_3$ 3:1, heating under 250 °C in heater plate so that each portion is added to dryness before adding a new portion. Then added 3 portions of 10 mL of $HClO_4$ 69-72% and 10 mL of 65% HNO_3 and brought to dryness, and finally were added 3 portions of 5 mL HF 48% and 10 mL of HNO_3 65% and brought to dryness. For elimination of $HCl:HNO_3$ 3:1, HF and $HClO_4$ were added 3 portions of 5 mL of HNO_3, H_2O_2 and 2 mL of 2 mL of deionized water. The samples were then completely dried, cooled and the salts were dissolved in about 20 mL of HNO_3 8M and then transferred to 100 mL volumetric flask and completed with the same solution. Volumetric flask were removed and transferred to glass vials, 10 mL aliquots for each sample.

2.3. Determination of the Activities of Radionuclides

To determine the activity concentration of a sample by gamma spectrometry before it is necessary to perform the calibration function of the energy efficiency of the equipment. For

this work, the calibration curve was constructed using a mixed source in liquid form, containing the following radionuclides: ^{241}Am, ^{152}Eu, ^{137}Cs and ^{60}Co, which have photopics in energy: 59.54; 121.78; 661.66 and 1332.49 keV, respectively.
A sample of the background (BG) was prepared and analyzed under the same conditions of the samples, this result being subsequently subtracted from the results obtained for each filter sample.
The filter cartridge samples were analyzed by gamma spectrometry. Aliquots of about 10 mL were then sealed in vials with calibrated geometry for gamma spectrometry measurements in order to determine the activity concentrations.

2.4. Analytical and Nuclear Techniques

A Gamma Spectrometry System (Model GX 2518 from Canberra Industries) was employed in this study, consisting of a HPGe (High Purity Germanium) detector.

3. RESULTS AND DISCUSSION

The system efficiency of detection depends on gamma radiation energy, thereby adjustments were made in the system, energy and efficiency. Figure 2 shows the calibration curve as a function of energy efficiency. It is necessary that a calibration curve is made for each detection system and used the counting geometry.

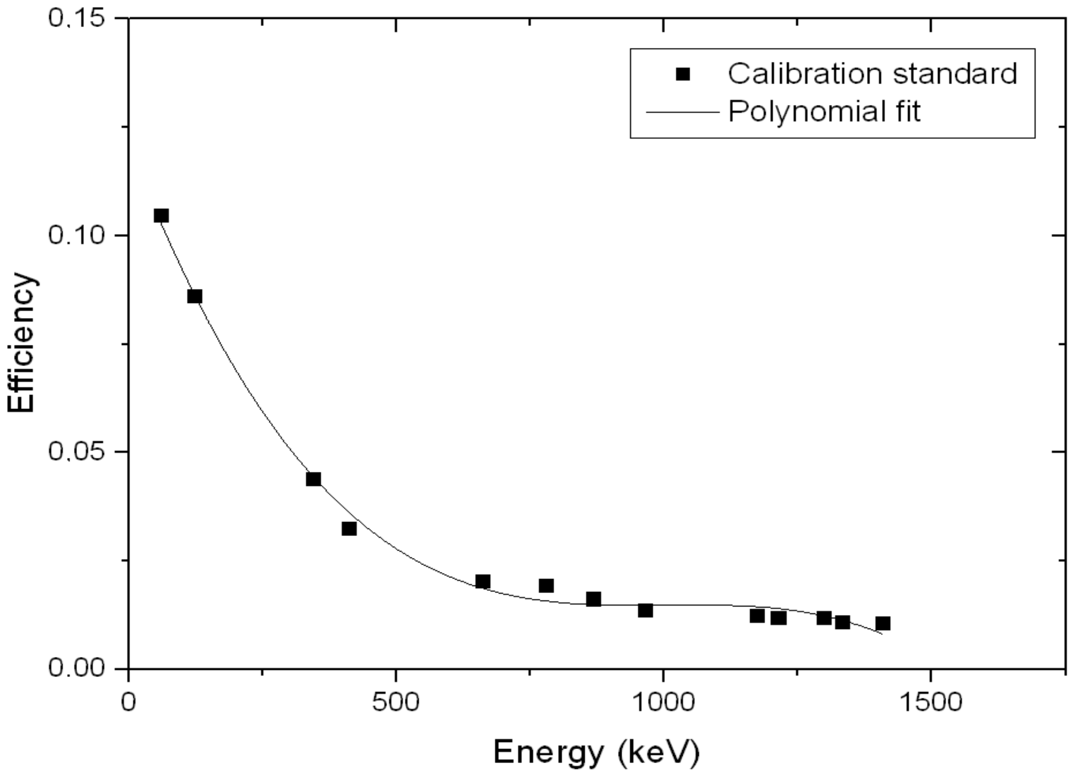

Figure 2: Calibration curve efficiency in energy function

The samples were analyzed by gamma spectrometry, allowing to qualify and quantify the present radionuclides. The spectrum shown in Figure 3 demonstrates radionuclides which have been identified in the sample. All filter cartridges analyzed to date show these same radionuclides, namely: ^{108m}Ag, ^{110m}Ag and ^{60}Co.

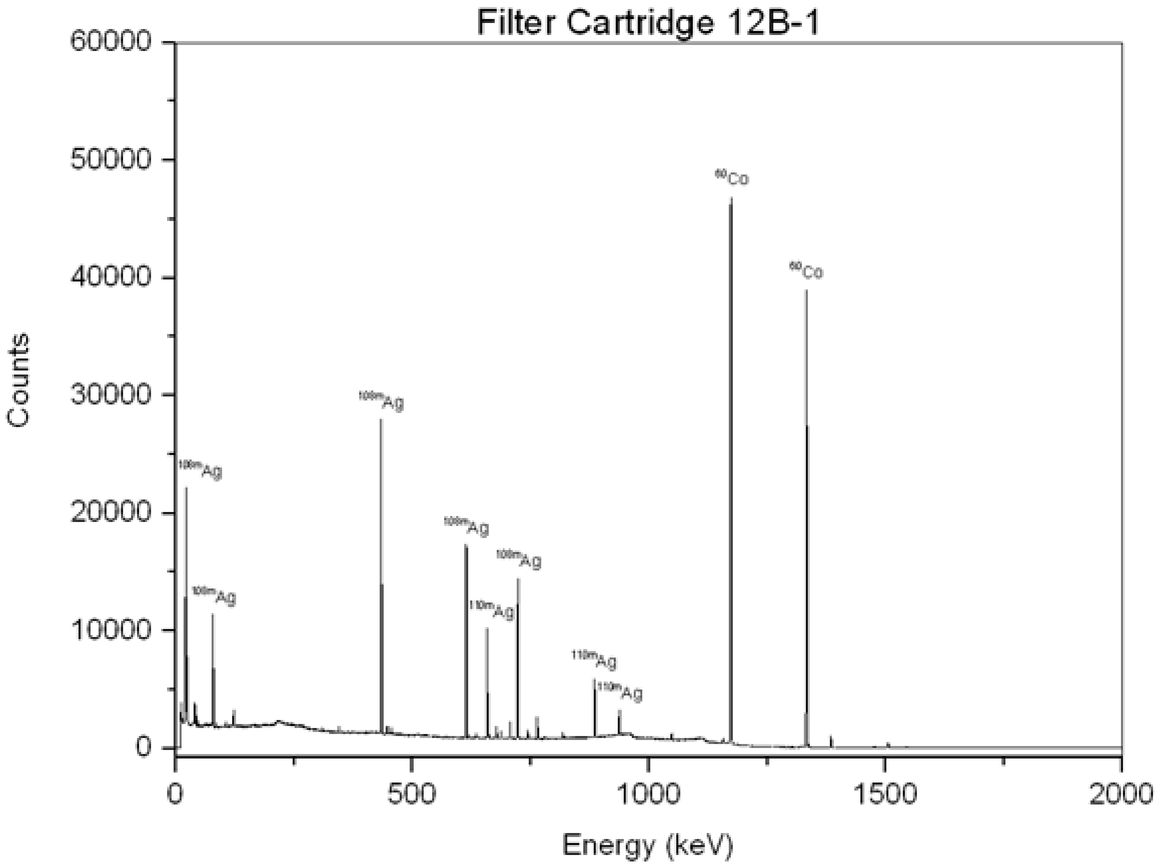

Figure 3: Spectrum obtained on the sample filter cartridge 12B-1

The concentration of activity was calculated from Equation 1, where:

$$A = \frac{Ap}{\gamma . T\varepsilon(E)} \quad (1)$$

A - Activity (Bq);
Ap - Net peak area;
γ - Probability of gamma emission;
T - Time count (s);
ε - Theoretical Efficiency.

Analyzing the results obtained so far with the help of Equation 1, it was possible to estimate total activity present in the filter cartridges 9.4 10^6 Bq order.

4. CONCLUSIONS

The method of radiochemical analysis developed for gamma emitting radionuclides was a laborious procedure, since involved calcination and dissolution steps. This procedure seems to be effective in characterization of the filters, but more samples must be analyzed to a final conclusion. The results obtained so far confirm the results presented by the authors Costa, 2015 and Tessaro, 2015. In these works the radionuclides found coincide with those found by the method presented in this paper and activity values are of the same order of magnitude.
This work is of great contribution to the development of characterization of radioactive waste program conducted in the laboratory of the management of radioactive waste of the IPEN.

ACKNOWLEDGMENTS

The authors would like to thank the National Nuclear Energy Commission / Nuclear and Energy Research Institute (IPEN-CNEN/SP) for an institutional grant.

REFERENCES

1. Instituto de Pesquisas Energéticas e Nucleares. *(Folheto fornecido aos visitantes para visitas técnicas). Reator IEA-R1. Divisão de Comunicação Social, IPEN*. São Paulo, (2002).

2. G. Hiromoto, J. C. Dellamano, J. T. Marumo, L. S. Endo, R. Vicente, T. Hirayama, Introdução à gerência de rejeitos radioativos. *Instituto de Pesquisas Energéticas e Nucleares - IPEN, Departamento de Rejeitos Radioativos – GRR*, São Paulo, (1999).

3. Comissão Nacional de Energia Nuclear – CNEN. Critérios de Aceitação para Deposição de Rejeitos Radioativos de Baixo e Médio Níveis de Radiação. *CNEN-NN-6.09.* Rio de Janeiro, (2002).

4. Us Nuclear Regulatory Commission. Licensing Requirements for Land Disposal of Radioactive Waste. *Code of Federal Regulations*, **Title 10, Part 61, Federal Register 47,** p. 57446, December, (1982).

5. T. Kekki, A. Tiitta, Evaluation of the radioactive waste characterization at the Olkiluoto nuclear power plant. *Helsinki Radiation and Nuclear Safety Authority*. March, (2000).

6. International Atomic Energy Agency. Determination and Use of Scaling Factors for Waste Characterization in Nuclear Power Plants. *IAEA - TECDOC-1363*. Vienna (2009).

7. International Atomic Energy Agency. Strategy and Methodology for Radioactive Waste Characterization. *IAEA - TECDOC-1537*. Vienna (2007).

8. P. Costa, *Determinação de uma metodologia para caracterização do filtro cuno do reator*

IEA-R1 utilizando o método de Monte Carlo. Dissertação de Mestrado – Instituto de Pesquisas Energéticas e Nucleares – IPEN-CNEN/SP, São Paulo (2015).

9. A. P. G. Tessaro, R. Vicente, Characterization of Spent Filters from the Water Polishing System of IEA-R1 Reactor. *Waste Management Conference – WM 2015*. Phoenix, Arizona, USA, March, 15 to 19 (2015).