



Methodology for filters analyzing by acid extraction

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1. Introduction

Currently, the use of nuclear technology has promoted significant developments in the use of nuclear technology in various sectors of society, such as scientific research, food preservation, electricity generation and the diagnosis and treatment of diseases. All these applications have brought many benefits to society [1].

However, the growing use of nuclear technology has prompted attention to the management of radioactive waste regarding contamination levels, in order to keep them as low as they do not compromise human well-being.

The National Nuclear Energy Commission (CNEN) establishes criteria for the evaluation and safe disposal of these wastes, with characterization being the most important criterion. The common methods of characterization are chemical attack and abrasive attack, but these generate by-products which could then become other radioactive waste [2].

Laser ablation is an option that has been tested for the treatment and characterization of radioactive waste at the Nuclear Energy Research Institute (IPEN) are developing a laser ablation probe to decontaminate surfaces. The current configuration involves a laser beam incident on a contaminated surface, followed by the exhaustion of the ablated material, which is retained on paper filters [3].

The material retained on the filters serves as a sample for characterizing the radioactive waste, fulfilling one of the criteria established for the safe disposal of radioactive material. For this reason, it became necessary to carry out a study to evaluate the chemical extraction yield of different filters, in order to identify which filter is ideal for use in the laser ablation probe.

Calcination and total dissolution in acids are the two most used methods for filter analysis. The calcination method is limited due to the volatility of some elements, causing considerable loss of sample for analysis. Total dissolution of the filter with acid, on the other hand, is impractical for physically resistant filters [4].

For this reason, this work aims to evaluate the recovery yield of the radioactive elements Cs, Co, U and Th using the hot acid extraction method. To compare the recovery yields, different types of filters were tested.

2. Methodology

2.1 Materials

For the extraction tests, standards of ^{60}Co , ^{137}Cs , ^{238}U and ^{232}Th were used. These radionuclides are commonly found in radioactive and nuclear installations. The ^{60}Co , ^{137}Cs standards were diluted in nitric medium and had the following activity concentrations, respectively: 789.75 Bq/g, 816.64 Bq/g. The ^{238}U and ^{232}Th standards were diluted in 2% nitric acid and had a total concentration of 1000 $\mu\text{g/mL}$. During the course of the work, five filters from different manufacturers were tested for extraction. It is shown in Table 1 the filter specifications: porosity, diameter and composition.

Table I: Specifications of the filters used in the chemical extraction tests.

Filter	Manufacturer	Porosity (μm)	Diameter (cm)	Composition
Type I	GelmanScienciens	0.8	25	Cellulose membrane
Type II	Axiva	1.6	47	Filberglass
Type III	Millipore	0.8	47	Glass microfiber
Type IV	Millipore	0.2	47	Polyethrsulfone membrane
Type V	Millipore	3.0	47	Cellulose esters

2.2 Extraction test on hot plate and activity measurement

For the initial concentration activity in each filter, 20 μL of the ^{60}Co , ^{137}Cs , ^{238}U and ^{232}Th standards were pipetted onto the filters paper (Table I). The resulting activity was 15.79 Bq/filter for ^{60}Co , 16.33 Bq/filter for ^{137}Cs , 250 Bq/filter for ^{238}U and 81 Bq/filter for ^{232}Th . Three samples were prepared for each filter. The filters were then dried in an infrared lamp and counted using gamma spectrometry and neutron activation analysis. In the extraction procedure, after pipetting the same amount of each radionuclide in the filters in triplicate, they were putted in separate beakers, which were then filled with 50 mL of 3 mol L^{-1} HNO_3 and placed on a hotplate at a temperature of 80 degrees. The beakers were closed with watch glass and the procedure lasted 2 hours.

Once the extraction procedure was complete, the extracting liquid was filtered and poured into a beaker, placed on a hotplate at 80 $^{\circ}\text{C}$, the volume was reduced to approximately 0.5 mL and poured into plastic containers. The container was then placed under infrared light so that the volume of the extracting liquid was reduced near to dry.

A Whatman filter paper with 1.2 μm pores, with the same dimensions as the one initially containing the solution, was prepared and placed over the remaining liquid in the plastic container so that it absorbed it completely in order to return the sample to its original geometry. The filter paper in the plastic container was left to dry completely so that gamma spectrometry, for ^{60}Co , ^{137}Cs , and neutron activation analysis, for ^{238}U and ^{232}Th determination could be carried out. The ^{238}U determination was carried out through short- and long-term neutron activation procedures. The sample supports were also dried and measured after extraction.

For determination by instrumental neutron activation analysis, the filters were irradiated by 20 s in the short-term and for 8 hours in the long-term irradiation, in a thermal neutron flux of 1.0×10^{12} n cm^{-2} s^{-1} in the nuclear

research reactor IEA-R1 at IPEN-CNEN/SP. Primary standards were simultaneously irradiated with the samples for standardization and quality control purposes.

Gamma ray measurements were performed using a EG&G Ortec HPGe detector coupled to a multichannel analyzer. The spectra were analyzed using gamma analyst and CAX software [5] both for gamma spectrometry and neutron activation analysis.

3. Results and Discussion

During the chemical extraction stage, it was noticed that the filters differed in their degree of dissolution. The Type I filter dissolved completely, filters II, III and V dissolved partially, while filter IV did not dissolve. When it comes to chemical extraction, it is important that the filter does not dissolve.

Table 2 shows the chemical yields of the radioisotopes analyzed in this work. The filters containing ^{60}Co and ^{137}Cs had chemical yields of over 80% with satisfactory data reproducibility. The type I filter, which was completely dissolved, has a chemical yield close to the type V filter, which does not dissolve completely.

Table 2: Chemical yield in the five filters tested in hot acid extraction in % and standard deviation (n=3) of recovery.

Filter	^{60}Co %	^{137}Cs %	^{238}U % (20 s)	^{238}U % (8 h)	^{232}Th %
Type I	88±9	85±9	74±18	77±15	73±21
Type II	80.22±0.04	88±2	59±18	53±12	64±17
Type III	88±6	85.5±0.2	51±1	43±5	66±9
Type IV	104*	90±8	69±24	56±18	93±13
Type V	88±4	89.3±0.6	75±19	93±9	102±18

* Only one measurement

There is a study suggesting that the Type II filter should not be extracted with H_2SO_4 , since tests carried out obtained yields of 65% [6]. For the extraction technique proposed in this work with $3 \text{ mol L}^{-1} \text{HNO}_3$, a chemical yield of 87% was obtained. Type I and Type V filters have the higher chemical yields for the ^{238}U and ^{232}Th radioisotopes. Type II and IV filters have yields close to 50% for ^{238}U . For uranium and thorium analysis, the reproducibility was lower than for cobalt and cesium and the reason for this results should still be investigated.

4. Conclusions

This work proposed a simple methodology for analyzing filters using hot acid extraction as a substitute for total dissolution or calcination. It was observed that filters show different behavior in the face of acid dissolution: complete, partial or non-dissolution during the extraction process. The tests carried out with different types of filters showed that, for all brands, the chemical recovery yields were higher than 80% for ^{60}Co and ^{137}Cs . The chemical recovery yield of the extraction process of filter samples containing U and Th, determined in long- and short-duration irradiation, had low reproducibility. Type I and V filters are the filters that presented the highest recovery yields when it comes to all the radioisotopes analyzed. The extraction

process for activity concentration determination, in filters containing radionuclides, proved to be an efficient process. Nevertheless, the filter behavior must be precisely known and the recovery for that particular filter type must be determined before using the extraction process.

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