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Evaluation of recovery yields of ^{60}Co and ^{137}Cs in extraction procedures

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ABSTRACT

Nuclear technology has been implemented in several sectors of society, among them, it is possible to highlight medicine, industry, research, among others. The high demand of these nuclear applications has caused problems with regard to the generation of radioactive waste. The laser ablation method has been pointed out in the last years for the decontamination of surfaces and the contamination removed is completely retained in paper filters. This requires a safe and inexpensive method to perform the characterization of the radioisotopic inventory present in the laser filter samples. In this study, the extraction and quantification of the radionuclides present in the filter was evaluated using ^{60}Co and ^{137}Cs radionuclides. Two forms of extraction were tested: with Soxhlet equipment and by digesting the filter samples in a beaker, both using 3M HNO_3 as extractant. The chemical yield was calculated using gamma ray countings obtained before and after extraction. It was observed that the extraction method by digesting the filter directly in a beaker presented the highest yield recovery results.

Keywords: **nuclear technology, radiochemical characterization, radionuclide extraction.**

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1. INTRODUCTION

Nuclear energy is an area of science in continuous progress with a broad field of applications such as electric energy generation, diseases diagnosis and treatment, quality control, and several other research areas which benefit the whole society [1]. On the other hand, these activities interfere in a certain way with the environment. Radioactive materials used and the waste generated in these applications can be harmful to life. Currently, one of the major problems in this sector is the radioactive waste from radioactive or nuclear installations.

The Laser Center and Applications (CELAP) and Radioactive Waste Management (GRR) of Instituto de Pesquisas Energéticas e Nucleares (IPEN) are committed to developing a laser ablation technique as a mean of treatment for contaminated surface materials, aiming its further disposal as radioactive waste or recycling [2]. This probe removes the surficial layer of contaminated material, which is ejected and converted into solid aerosol. Then, the aerosol formed is retained in a HEPA filter.

To assess the efficiency of surface contaminants removal by the laser, a radiochemical analysis of the filter present in the laser ablation probe is necessary to determine the radioisotope inventory of the sample [3].

One of the filter methods of analysis is the dissolution, where the filter is completely dissolved using a combination of acids [4, 5]. However, performing dissolution of filters is an extremely laborious technique, due to the fact that the filters used in these activities are chemically stable for a variety of acids. Thus, the dissolution method may sometimes become unfeasible even using hot acid combinations [6].

Another method of analyzing filters is the calcination, in which the sample is completely burned in an oven. Zapata et al. tested the method on filters, which were placed in a baffle furnace at a temperature of 450 °C, the residual ash being completely digested in acids [5]. However, the calcination technique is limited for some radionuclides due to the possibility of sample loss.

The objective of this work was to evaluate a quick and simple methodology for ^{60}Co and ^{137}Cs radioisotopes extraction present in filters. Two methods for extraction of these radionuclides trapped in the filter samples were tested. The first one was by using Soxhlet extraction equipment

and, the second consists in digesting the filter in acidic solution in a beaker. In both cases 3M HNO_3 was used as the extractant. The radioisotopes ^{60}Co and ^{137}Cs were chosen due to the fact that they are commonly found in radioactive and nuclear installations [7].

2. MATERIALS AND METHODS

2.1. Materials and reagents

All chemical reagents used in this work were of analytical grade. Standard solutions of ^{60}Co and ^{137}Cs were purchased from the Institute of Radioprotection and Dosimetry IRD/CNEN and diluted in nitric medium to present an activity concentration of 790 Bq g^{-1} and 817 Bq g^{-1}, respectively. From these solutions, 20 µL were taken and pipetted in micro filters with 1.2 µm pores for the extraction tests. The volume of 20 µL of the diluted solution provided a safe activity for handling, and counting statistics with the necessary precision for the measurements. In order to obtain the recovery of the process, the filters pipetted with the ^{60}Co and ^{137}Cs solutions were placed in a plastic support, counted by gamma spectrometry and the obtained counts per second taken as the initial activity.

2.2. Extraction procedure

For Soxhlet extraction, two types of thimbles were used, cellulose and fiberglass, and the system was attached to a siphon tube. A condenser was fitted next to the tube. For extraction, 200 mL of 3M HNO_3 was used. The extracting liquid was recovered in a round-bottomed flask. The Soxhlet equipment is shown in Figure 1.

During the extraction procedure the heating mantle was kept at a temperature of 100 °C. Extractions were performed at two different time intervals: 1 hour, 1 hour more adding a new extracting volume of 200 mL of 3M HNO_3 and at 2 hours straight with the same extracting solution volume.

Once the extraction procedure was completed, the extraction liquid was placed into a beaker and taken to the hot plate at 80 °C. The volume was reduced near to dryness and transferred to a blank paper filter to keep the initial counting geometry.

The second extraction method consisted in adding the filter containing ^{137}Cs and ^{60}Co in a beaker with 50 mL of 3M HNO_3, covered with a watch glass, and heated on a hot plate at a temperature of 80 °C for four hours. This procedure was divided into two stages, the first extraction was carried out

for two hours, then the solvent extractor was transferred to another beaker. The procedure was repeated on the same filter for another two hours. The acidic solution of the second step was then added to the first one for analysis. The final volume was reduced near to dryness and transferred to a blank paper filter to keep the initial counting geometry for gamma spectrometry.

Gamma spectrometric counting was performed with a GC2018 Canberra HPGe detector coupled to a Canberra DSA-1000 multichannel analyzer. The counting time was 30 minutes. Spectral analysis was performed using gamma analyst and CAX software. The gamma spectrometry was applied to the dried filters containing the samples in the same plastic supports used before. The plastic supports without the filter and the Soxhlet thimbles were also measured after the extraction.

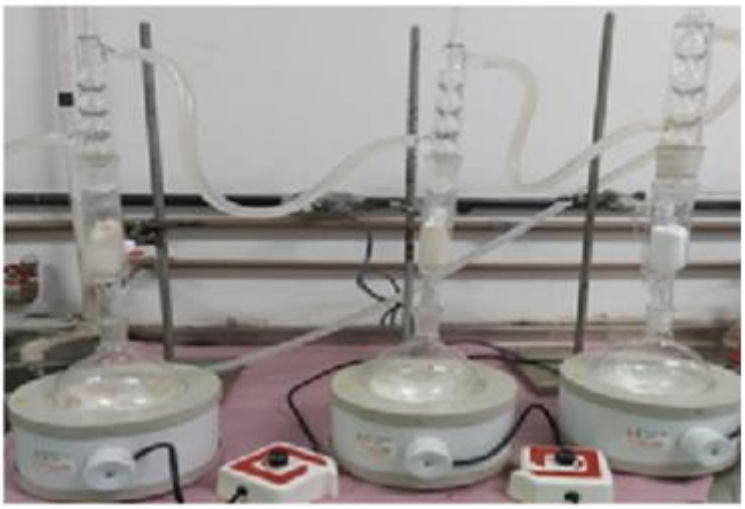

Figure 1: *Soxhlet equipment containing the samples for radionuclides extraction*

3. RESULTS AND DISCUSSION

The recovery yields of ^{60}Co and ^{137}Cs extracted from the filter for one hour with Soxhlet equipment showed unsatisfactory results. For this reason, the samples went through a second extraction process for another one hour, adding new 200 mL of 3M HNO_3. In the test, lasting 2 hours straight, without adding the new aliquot of extractant, the yields obtained were greater than for 1 hour, but lower than two extraction steps. The results are shown in Table 1.

The measurements of the plastic supports showed no counting in the range of energy transition decay of ^{137}Cs or ^{60}Co. On the other hand, it was noticed that part of the radionuclides was trapped on the thimbles, as shown in Table 2. The highest amounts of ^{60}Co and ^{137}Cs found for sample 3

must be due to the composition of the thimbles, given that those used for samples 1 and 2 were composed of cellulose, while the one used for sample 3 was composed of fiberglass (Figure 2).

Adding the yields obtained in the analyses of the thimbles with the extraction results for the three filter samples, the following values were obtained: 68% for the extraction of ^{60}Co and 55% for the extraction of ^{137}Cs indicating that part of the analyzed nuclides was also lost during the process.

Table 1: Chemical yield (%) of radionuclides analyzed by the two different extraction methods

	Soxhlet extracion for different time intervals						Extraction by acidic digestion of 4 h	
Sample	1 h	+ 1 h	2 h	1 h	+ 1h	2 h		
	^{60}Co			^{137}Cs			^{60}Co	^{137}Cs
1	53	71	68	55	65	61	98	115
2	54	86	81	67	78	75	84	101
3	45	61	40	35	46	7	92	106
Mean	51	73	63	52	63	48	91	107
Standard deviation	5	13	21	16	16	36	7	7

Table 1 also shows the mean value and standard deviation for the three repetitions of the experiment obtained using Soxhlet and beaker digestion extraction methods. On average, recoveries were lower in terms of chemical yields with larger standard deviation for the Soxhlet procedure when compared with the beaker digestion, for which the results were all satisfactory. Virtually all cesium and almost all cobalt were extracted.

These results indicate that the acidic extraction in the beaker provides higher chemical yield and reproducibility.

Evaluating the activity concentrations remaining in each support used in Soxhlet procedure, it can be seen that the cellulose thimble is less susceptible to adsorption of the radionuclides evaluated in this work than the fiberglass one (Table 2).

Table 2: Chemical yield (%) of radionuclides in the cellulose (1 and 2) and in fiberglass (3) thimbles

T	Chemical yield (%)	
	^{60}Co	^{137}Cs
1 (Cellulose)	4	1
2 (cellulose)	7	4
3 (fiberglass)	28	48

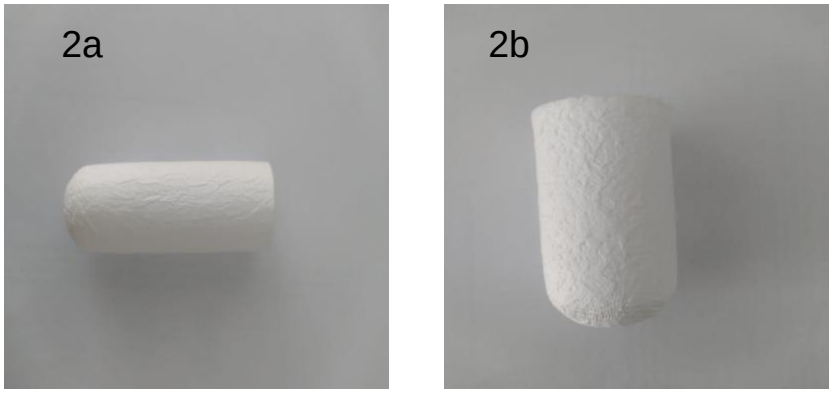

Figure 2: *Cellulose thimble used for extraction of samples 1 and 2 (2a) and fiberglass thimble used for extraction of samples 3 (2b).*

4. CONCLUSIONS

The ^{60}Co and ^{137}Cs extraction from filter samples using Soxhlet equipment and acidic digestion in beaker showed that the first one had chemical yields varying from 61 to 86% for cobalt, and from 46 to 78% for cesium in extractions procedures carried out in two steps of one hour. The fiberglass thimble used with sample 3 provided the lowest yield and a considerable amount of ^{137}Cs were retained in it. The extraction of filters containing ^{60}Co and ^{137}Cs by digestion in a beaker on hot 3M HNO_3 showed higher recovery yield results without considerable loss, mainly for cesium.

For the Soxhlet extraction procedure, it was observed that the time of extraction, amount of solvent and addition of more solvent can influence the recovery yields. Future tests varying these parameters must be performed in order to better understand the influence of these parameters.
It can be observed that the extraction by acidic digestion in a beaker for 4 hours on a hot plate is more efficient for the extraction of ^{60}Co and ^{137}Cs from filters. This work proposed a simpler methodology for filter analyses other than its total dissolution or calcination.

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