



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

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1. Introduction

Nuclear energy is an area of science in continuous progress, with a broad field of applications. Its applications such as electric energy generation, diseases diagnosis and treatment, quality control, and several other research areas benefit the whole society [1]. On the other hand, these activities interfere in a certain way with the environment. Radioactive materials used in these applications can be harmful to life. Currently, one of the major problems in this sector is 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 surface treatment for contaminated surface materials, for 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 to determine the radioisotope inventory of the sample is necessary [3].

One of the filter method of analysis is the dissolution, where the filter is completely dissolved using a combination of acids [4, 5]. However, performing dissolution in filters is an extremely laborious technique, due to the fact that 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, in addition to the possibility of sample loss.

The objective of this work was to evaluate a quick and simple methodology for radioisotopes extraction in filters. Two methods for carrying out the extraction of radionuclides present in the samples were tested. One using a Soxhlet equipment and the second consisting in digesting the filter in HNO_3 in a beaker. The radioisotopes analyzed in this work were ^{60}Co and ^{137}Cs , elements that are commonly found in radioactive and nuclear installations [7].

2. Methodology

2.1. Materials and reagents

All chemical reagents used in this work present 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 789.75 Bq g^{-1} and 816.64 Bq g^{-1} , respectively. From these solutions, $20 \mu\text{L}$ were taken and pipetted in micro filters with $1.2 \mu\text{m}$ pores for the extraction tests. 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, filter samples were added to a thimble of cellulose and other of fiberglass attached to a siphon tube. A condenser was fitted next to the tube. For extraction, 200 mL of HNO_3 was used. The extracting liquid was recovered in a round-bottomed flask. The Soxhlet equipment is shown in figure 1.

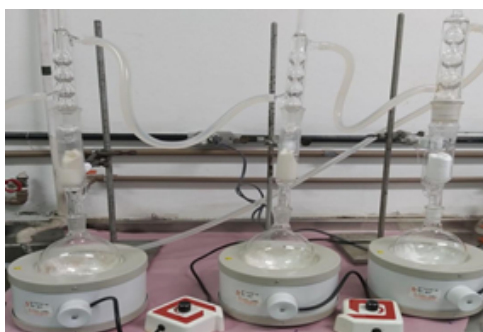


Figure 1: Equipment Soxhlet containing the samples for extraction.

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

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

The other 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 clock glass, and heat it on a hot plate at a temperature of 80°C for four hours. The extractant acid was separated and the process repeated. After that, the extraction liquid was dried to 0.5 ml and transferred to a paper filter for gamma spectrometry counting.

Gamma Spectrometric counting were 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.

3. Results and Discussion

The yield recovery of ^{60}Co and ^{137}Cs extracted from the filter for one hour with Soxhlet showed unsatisfactory results. For this reason, the samples went through a second extraction process for another hour, adding new 50 mL of 3M HNO_3 . In the test of 2 hours straight, without adding the new aliquot of extractant, the yields obtained were greater than for 1 hour, but lower than for the two steps extraction (Table I).

The measurement of the plastics support showed no counting for cesium or cobalt. On the other hand, it was noticed that part of the radionuclides was trapped on the thimbles, as shown in Table II. The higher amount of ^{60}Co and ^{137}Cs found in sample 3 must be due to the composition of the thimbles, given that those ones used in samples 1 and 2 were made up of cellulose, while the one in sample 3 were made up of fiberglass (Figures 2a and 2b) that is from a different brand from the ones used for samples 1 and 2.

Table I: Chemical yield (%) of radionuclides analyzed at different time intervals.

Sample	^{60}Co			^{137}Cs			Becker	Becker
	1 hour	+1 hour	2 hours	1 hour	+1 hour	2 hours	^{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

The samples in beakers had a duration of 4 hours.

Table II: Chemical yield (%) in samples of thimble of cellulose

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

Adding the yields obtained in the analysis of the thimbles with the extraction results for 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.

For the extraction tests carried out after cooking the filters in HNO_3 , the yields were all satisfactory (Table I), virtually all cesium and almost all cobalt was extracted.

4. Conclusions

The filter extraction test of ^{60}Co and ^{137}Cs samples using the Soxhlet equipment showed efficiency varying from 61% to 86% for cobalt and for cesium, varied from 46% to 78% with the extraction in two steps of one hour. The thimble used for sample 3, provided the lowest yield and a considerable amount of ^{137}Cs were retained in it. The extraction on filters containing ^{60}Co and ^{137}Cs by digestion in hot HNO_3 3M, showed high rates of recovery yield, without considerable loss mainly for cesium. For the Soxhlet procedure, it was observed that the time of extraction, amount of solvent and changing the solvent influence the recovery yield. Future tests varying these parameters must be done in order to get better results.



(a) Cellulose thimbles for samples 1 and 2



(b) Fiberglass thimble for sample 3

Figure 2: Thimbles used for extraction in filter

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