Determination of Scale Factor for Ni-59 and Ag-108m in Ion Exchange Resin From Angra 1

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1.Introduction

In Brazil, while the National Nuclear Safety Authority (ANSN) isn't fully operational, the National Nuclear Energy Commission (CNEN) is the authority responsible for establishing rules for the proper destination of nuclear waste in nuclear power plants, and the CNEN-NN-6.09 norm provides criteria for its correct disposal. For the proper destination, an inventory of all radionuclides present in each unit of radioactive waste must be well known, and among those the Difficult to Measure Radionuclides (DMRs) are a special case, because their determination is a non-trivial task in virtue the following matters [1]: i) absence of emission of gamma or X rays; ii) very low emission energy and/or intensity of the gamma/X rays; iii) low activity; etc. For the determination of these DMRs in nuclear waste from nuclear power plants, like Angra 1 and 2, the use of radiochemical separation techniques, which take long periods of time, is usually required [2]. The International Atomic Energy Commission – IAEA – recommends the use of indirect methodologies to determine the presence of DMRs in radioactive waste from nuclear power plants, among which the use Scale Factors (SF) [3]. In this methodology, the specific activity of a DMR is determined using a radioisotope that's easy to determine, called "Key Radionuclide" (KR), as comparator.

In this study the SFs of DMRs gamma emitters ^{59}Ni and ^{108m}Ag were determined by gamma spectroscopy using ^{60}Co as KR in 12 samples of ion exchange resin nuclear waste from Angra 1 nuclear power plant.

2. Methodology

2.1 Scale Factor

The Scale Factor method can be used when, in a given matrix, a relationship is found between a DMR and a nuclide which can be easily determined, the KR – this may not be possible for some radioisotopes and the Scale Factor (SF) is usually matrix-dependent. In general terms, the SF is the ratio between the concentration of the DMR and that of the KR:

$$SF = \frac{A_{DMR}}{A_{KR}}$$

(1)

where A_{DMR} is the specific activity of the DMR and A_{KR} is the specific activity of the KR. In order to verify if the SF method can be used for that DNR/KR pair in a given matrix, and if so to determine its value for posterior use, the concentrations of the DNR and of the chosen KR have to be determined in a set of individual samples from a specific type of matrix; then, as in some cases the activities and/or the SF may differ by orders of magnitude, a geometrical mean is used to obtain a working value [1]:

$$\overline{SF} = e^{\left(\Sigma_{i=1}^{N} \ln (SF)_i \middle/ N \right)} \tag{2}$$

where N is the number of individual samples. In order to the SF methodology be considered acceptable for that DNF/KR pair in that specific matrix, the maximum variation in individual measurements of the SF must be comprised between a factor of 10 below or above the average, i.e. [1].

$$\frac{\overline{SF}}{10} \leq (SF)_i \leq \overline{SF} \cdot 10 \tag{3}$$

2.2 Separation of ^{59}Ni

After digested, 10 mL aliquots of ion exchange resin samples were used for the ^{59}Ni separation. Each aliquot was heated up to 70ºC and then 10 mL of dimethylglyoxime (DMG) 1% was added for Ni precipitation. Then, NH_4OH (0,1M) was added under stirring and then 1 mL of DMG was added in order to have total precipitation of Ni.

To guarantee the homogeneity and total fixation of Ni, the precipitate was filtered under vacuum using fiber glass filter; after drying, the filter was covered with parafilm. The determination of ^{59}Ni SF was performed by gamma spectroscopy using the 6.29 keV K_α X-ray.

To determine the separation yield of Ni, 2 mL of stable Ni were added in 5 mL aliquots of the original samples, then these were dried and recovered with 5 mL of HCl (1M). The separation yield was determined using Atomic Absorption Analysis.

2.3 Separation of ^{108m}Ag

For the ^{108m}Ag separation, aliquots with 40 mL of ion exchange resin from Angra 1 were used, to which 118.18 mg of $AgNO_3$ and HCl_{conc} were added in order to precipitate AgCl. Subsequently, the samples were inserted in Falcon tubes and centrifuged for 25 minutes under 3500 rpm. The precipitate was deposited in filter paper and then dried. The separation yield was determined by gravimetry, and the SF was determined by gamma spectroscopy using the 79 keV gamma transition from ^{108m}Ag.

2.4 Gamma Spectroscopy

A 40% Canberra XtRa Extended Range Coaxial HPGe was used for the gamma spectroscopy, and the spectra analysis was performed using Genie 2000® software. The efficiency correction was performed using Canberra® ISOCS/LABSOC, which uses Monte Carlo simulations to determine the efficiency [4].

3. Results and Discussion

Fig. 1 presents the variation of ^{60}Co specific activity on the 12 samples of ion exchange resin before any radiochemical separation, which show a good homogeneity, with all values in the same order of magnitude.
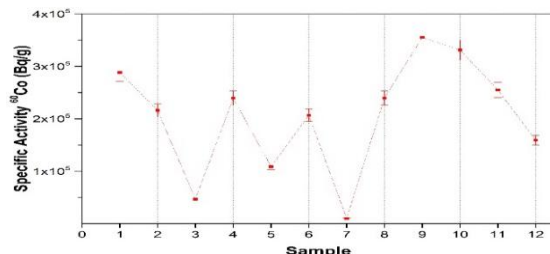
Figure 1: Specific activity of ^{60}Co in ion exchange resin from Angra 1.

On Fig 2, the spectra obtained for one of the samples are shown, (A) before any radiochemical separation, (B) after ^{108m}Ag separation, and (C) after ^{59}Ni separation.
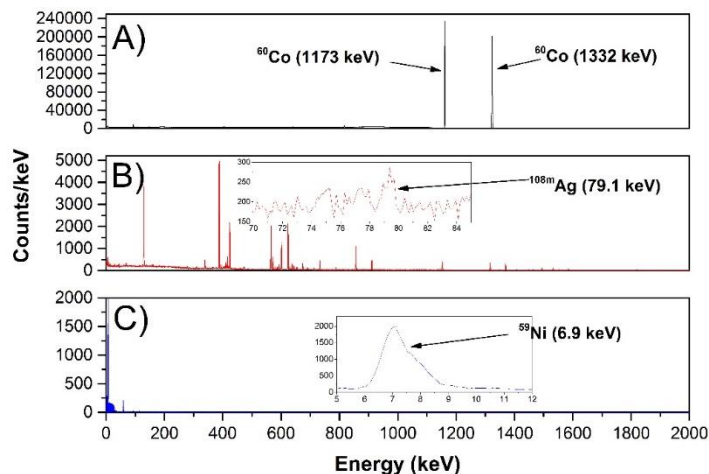

Figure 2: Spectra obtained for one of the samples, with a zoom in the peak of interest in the insert of each graph: **A)** before radiochemical separation; **B)** after ^{108m}Ag separation; **C)** after ^{59}Ni separation.

When the spectrum was analyzed before any separation, a strong presence of ^{60}Co was verified, whereas it wasn't possible to detect the presence of either ^{59}Ni or ^{108m}Ag as both were below the detection limit. However, after the radiochemical separations were carried out, a strong decrease in the amount of ^{60}Co was verified, and the presence of ^{108m}Ag and ^{59}Ni could be detected and quantified.

The individual SF results for ^{59}Ni and ^{108m}Ag for each of the 12 samples are presented on Figs 3 and 4, respectively, together with the mean value. These values obey the rule presented on equation 3, barring the SF obtained for ^{59}Ni on the aliquot #7, in which it was not possible to determine the presence of ^{59}Ni, even after the radiochemical separation, indicating that the concentration of this radioisotope in this sample is below the detection limit.
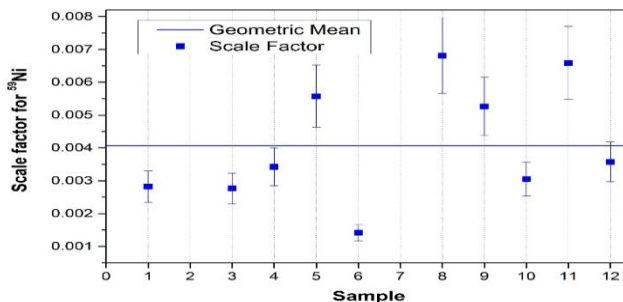

Figure 3: Distribution of the Scale Factor values for ^{59}Ni in ion resin exchange samples from Angra 1.
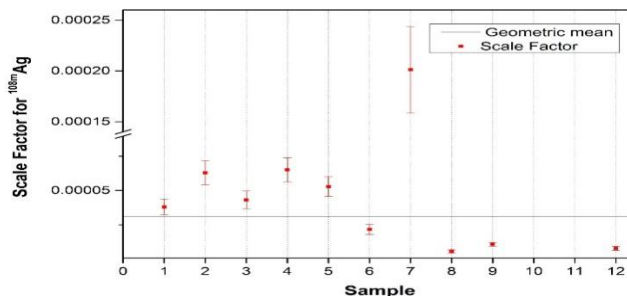
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Figure 4: Distribution of the Scale Factor values for [108m]Ag in ion resin exchange samples from Angra 1.

4. Conclusions

In this study the Scale Factors (SFs) for [59]Ni and [108m]Ag were determined in 12 ion exchange resin samples from Angra 1. The results pointed out a homogeneous distribution of ^{60}Co on the samples. Furthermore, the radiochemical separation methods applied proved to be suitable for the separation of these radioisotopes on the analyzed matrices. The SFs determined can be regarded as satisfactory for the analyzed samples and thus can be used as a starting point for the determination of [59]Ni and [108m]Ag in ion exchange resin wastes from Angra 1.

Acknowledgements

References

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