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DETERMINATION OF I-129 ON RADIOACTIVE WASTE FROM ANGRA 1 AND 2 POWER PLANTS

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ABSTRACT

Radioactive waste must be characterized in order to be deposited in a suitable place for its confinement, avoiding any contamination of the environment. This study determined ^{129}I, one of the difficult-to-measure nuclides that make up the radioactive waste from Angra 1 and 2 nuclear power plants. The methodology used a 129-I (1Bq/mL) liquid certified standard and consisted on guarantying that all the iodine was converted into iodide before the addition of $PdCl_2$ for the precipitation of PdI_2. After the separation procedure the ^{129}I was quantified by gamma spectrometry by measuring its X-ray emissions with energies of 29.7 keV and 33.8 keV, as well as the gamma-ray emission of 39.6 keV. The chemical yield of the procedure was 74.4% (0,2).

1. INTRODUCTION

Many iodine isotopes can be found in the radioactive waste of a nuclear reactor [1] and according to norm CNEN NN 6.09, which defines criteria for acceptance of waste radionuclides of low and medium activity for storage [2], all radionuclides that compose the waste must be quantified with enough precision to be within the authorized limits.

^{129}I is one of the most important and difficult to measure nuclides, important due to the long half life (15,700,000 years) and difficult to measure due to the low intensity of its only gamma emission (39.6 keV, 7.5%) [3] – the low energy of the gamma transition also requires the use of low-energy capable detectors.

This work focused on the study of the chemical processing required for the iodine precipitation and on the study of the measurement of ^{129}I.

2. MATERIALS AND METHODS

For these measurements, samples with 1, 5 and 10 mL of a certified liquid standard of ^{129}I (1 Bq/g) were diluted in 30 mL of 3M NaOH for iodine stabilization. Then, as the iodine amount was too low, for each sample 1mL of 0.0785M KIO3 was added as a carrier. To reduce iodate to iodine, 0.62 g of hydroxylamine hydrochloride was added and the pH of the solution was adjusted to lie between 1 and 2. The precipitation was accomplished by adding 1 mL of a PdCl2 solution, so that iodine was precipitated as PdI2. The solution was then filtered on

glass fiber paper and the iodine-deposited filters were placed in a petri dish and dried in a stove for 24h at 50°C. The samples were then covered in Parafilm® for protection.

The electromagnetic spectrum analysis was performed using a 40% Canberra XtRa Extended Range Coaxial HPGe, which has an operating range from 3 keV to >10 MeV [4], and the spectra analysis was performed using Canberra's Genie-2000 software [5]. The detection efficiency was calculated using Canberra's ISOCS/LABSOCS methodology [6], which performs a Monte Carlo simulation of the geometry. This way, the activity of each sample can be determined as:

$$A = C / (t_c \cdot \varepsilon \cdot I_\gamma) \quad . \tag{1}$$

where A is the sample activity (in Bq), C is the number of counts in the peak, t_c is the counting time, ε is the detector efficiency (obtained in the LabSOCS software) and Iγ is the transition intensity.

3. RESULTS AND DISCUSSION

The low energy spectra of ^{129}I are quite easy to fit, as the three peaks of interest (K_α at 29.7 keV, K_β at 33.8 keV and γ transition at 39.6 keV) are clearly separated (see Fig. 1). However, although the K_α line is the most intense one, in some cases it may be more interesting to use the gamma transition, to avoid possible interference from other iodine isotopes that may be present in a real-life sample. Table 1 shows the relevant data on each of these three emissions.

Table 1: Relevant data on the ^{129}I transitions of interest [7].

Transition	Energy (keV)	Intensity (%)
K_α *	29.7	58.1 (16)
K_β *	33.7	12.6 (3)
γ	39.7	7.51 (23)

*. The x-ray line energies were obtained by a weighted average of the individual sub-lines, and the intensity is the sum of the individual intensities

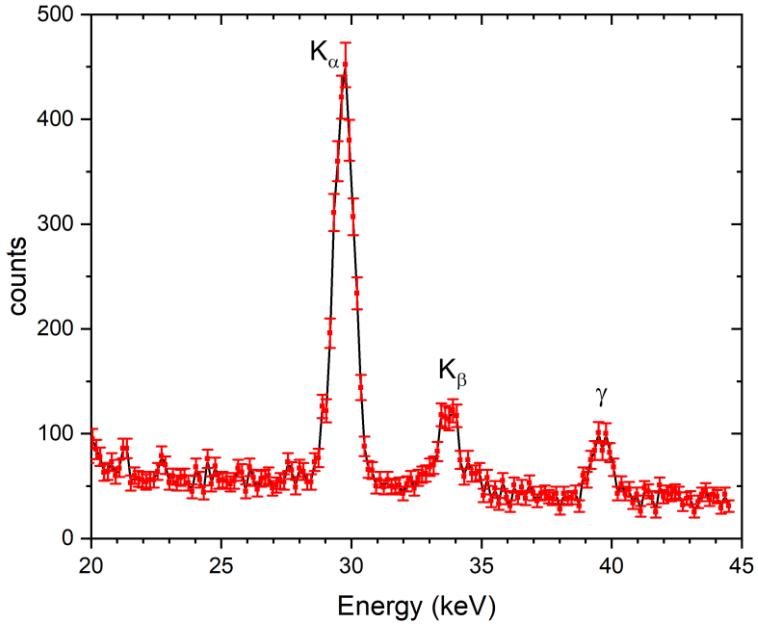

Figure 1: Low-energy spectrum from a ^{129}I sample.

The activity in each of the three samples was calculated, and the results are shown in Fig. 2; as the results were in perfect agreement within the uncertainties, the weighted average of these activities was calculated and is also shown in the figure.

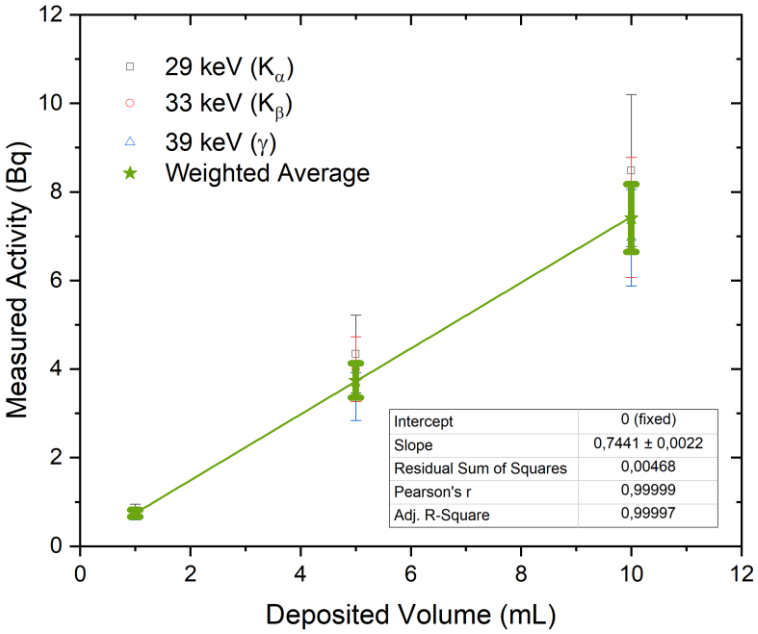

Figure 2: Measured activity of each sample as a function of the initial volume deposited in the sample – the green dots and line refer to the weighted average of the results for each line.

The fit of a linear function with the intercept fixed to zero (as no ^{129}I would be expected in a blank sample) resulted in a very good fit, with Pearson's r of 0.99999 and an adjusted R-Square of 0.99997, indicating the linearity of the activity with the volume of the sample (i.e., with the expected activity). As the original standard solution had an activity concentration of 1Bq/mL, the slope of the fitted function can be understood as a good estimative of the chemical yield of the deposition process – in this case, the result was (74.41 ± 0.22) % - further studies will be undertaken to assess the chemical yield by other methods and verify this result.

4. CONCLUSIONS

The analysis of the ^{129}I spectra showed that both x-ray transitions and the 39.6 keV gamma ray line are easy to separate in the spectra. The measured activity of the samples in the interval assessed by this study proved to be directly proportional to the deposited volume for all of the transitions, as well as for the average of the three of them, indicating that the activity of this radioisotope can be determined by either of these transitions. However, as in a real-life sample there could other iodine isotopes contributing to the x-rays, the 39.6 keV gamma ray transition may be a better choice. Additionally, from the initial activity and the measurement results, the chemical yield of the process was estimated in (74.41 ± 0.22) %.

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