



Development of a method to analyze chlorine-36 activity concentration in radioactive wastes from Nuclear Power Plant

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1. Introduction

Nuclear reactor activities, regardless of their purpose, generate radioactive waste whose management must be focused on mitigating potential damages to environment and human health [1]. Before being stored, the radioactive waste has to be characterized and classified in relation to their content, according to acceptance criteria defined by regulatory agencies [2]. In Brazil, the legislation 10.308/01 establishes the standards for final destination of radioactive waste produced in national territory. Final disposals are known as repositories, where the waste that is not intended to be removed in future is stored [3]. Furthermore, the CNEN NN 6.09 standard established by National Nuclear Energy Commission (CNEN) demands the characterization of low or medium-activity waste using analytical techniques, in order to prove their compliance to acceptance criteria for storage repositories [4].

Radioactive waste characterization is a complex task and gamma-ray spectrometry is usually applied to estimate the activity of high-energy and intense gamma emitters, which are considered “easy to measure”. However, radionuclides in nuclear waste emitting low energy gamma or X-ray, beta or alpha radiations are classified as “difficult to measure”, due to radiochemical separation requirement for their determination [5-6]. The radionuclides present in nuclear reactors waste can be formed as activation products (³H, ¹⁴C, ⁵⁴Mn, ⁵⁵Fe, ⁶⁰Co and ⁶³Ni); as fission products (⁹⁰Sr, ¹²⁹I, ¹³⁴Cs, ¹³⁷Cs, ¹⁵²Eu and ¹⁵⁴Eu); and uranium isotopes and transuranic elements ²⁴¹Am, ²⁴⁴⁺²⁴³Cm, ²⁴²Cm, ²³⁷Np, ²³⁸Pu, ²³⁹⁺²⁴⁰Pu, ²⁴²Pu and ²⁴¹Pu [7]. These radionuclides are frequently determined in low and medium activity waste. Nevertheless, the radionuclides ⁹⁴Nb, ⁹³Mo, ⁹⁹Tc and ³⁶Cl are equally important for characterization of radioactive waste from reactor operations.

The ³⁶Cl radioactive isotope is a beta particle emitter with a long half-life of 3.01 x 10⁵ years and is a neutron activation product from stable chlorine, which is present as an impurity in construction materials of the nuclear reactor facilities. The determination of this nuclide requires the complete separation of the matrix, due to interferences of other elements present in the sample, either due to its very low concentrations or because its radiation can be absorbed by the sample [8].

As mentioned before, the final disposal of nuclear facilities waste must have its radionuclide composition completely characterized as well as the concentrations of activities determined. This characterization requires the existence of a well-defined method, with sufficient precision and accuracy to the specific purpose. Therefore, this study aims to implement a fast method to determine ³⁶Cl concentration of activity in nuclear waste samples from nuclear centers.

2. Methodology

Prior to ^{36}Cl determination, nuclear waste samples were completely decomposed (Fig. 1), followed by radiochemical separation (Fig. 2). Chlorine was separated by AgCl^{36} precipitation and the dissolved precipitate was purified by ion exchange chromatographic column, in order to isolate the radioactive isotopes of Ag that could be present in the sample. The concentration of activity determination was carried out by Liquid Scintillation Counting (LSC) during 1h in triplicate measurements for each sample and two blank solutions, which were used to assess the background fluctuations [9]. LSCs were performed in the laboratory of IPEN's Radioactive Waste Management Service (SEGRR) using a HIDEX detector. The chemical yield determination was obtained using Gravimetric analysis of the chlorine stable isotope present in the sample, once the mass of hydrochloric acid added for the formation of AgCl^{36} precipitated is known. The chemical yield was between 89 and 104%.

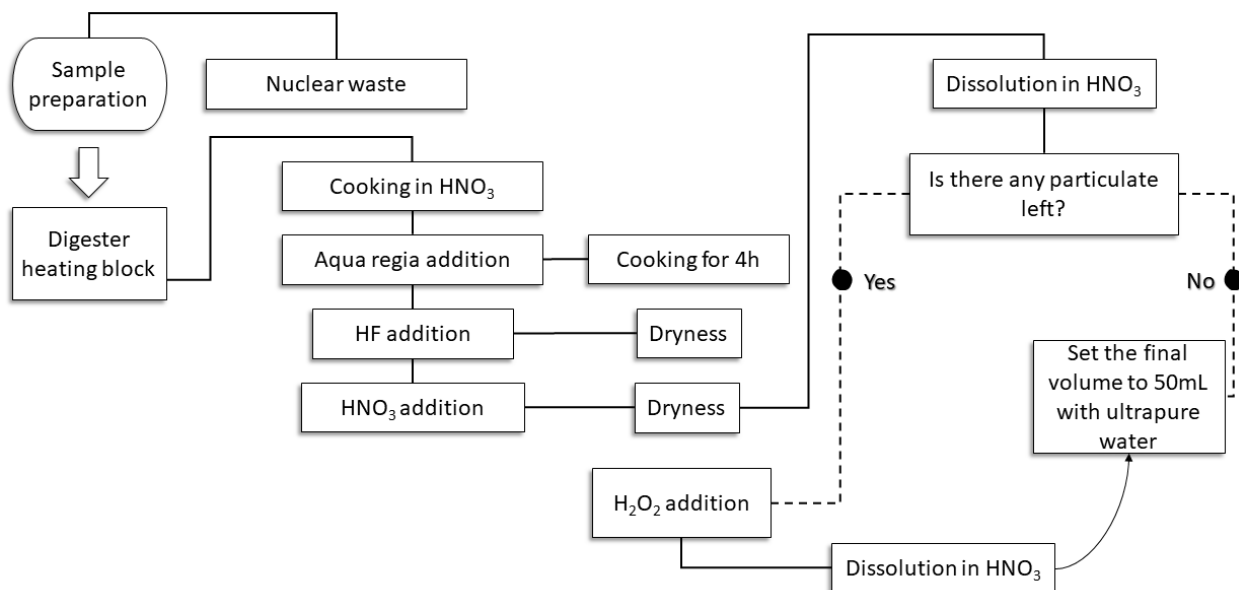


Figure 1: Decomposition method for nuclear waste samples.

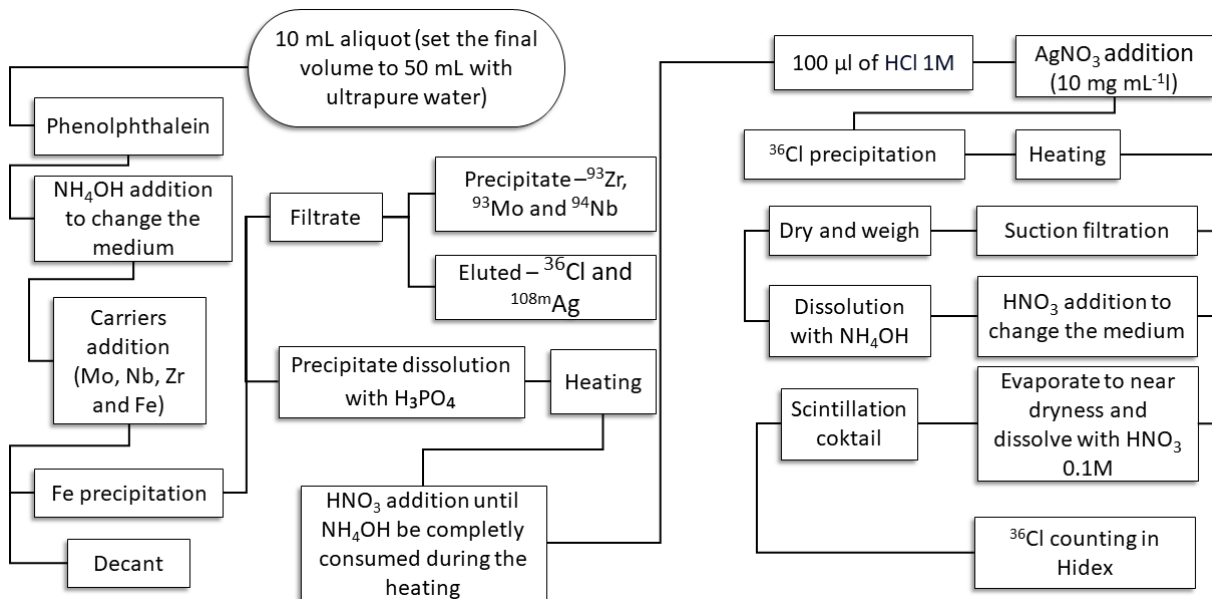


Figure 2: Radiochemical separation method for chlorine-36.

3. Results and Discussion

The results of ^{36}Cl analysis using LSC technique are presented in Table 1. ^{36}Cl activity concentrations were above Detection Limit (DL) for all analyzed samples, which indicates that the applied method for chlorine-36 determination in nuclear waste is suitable. Furthermore, it is important to emphasize that chlorine-36 is a difficult to measure radionuclide and the results obtained have a great importance, once this work is a proposal of a fast method development. Detection Limits (DLs) were estimated using confidence limits (3-sigma of BG values), representing a 99% confidence level that the event is real and not just a statistical fluctuation. The average and standard deviation ($n = 3$) for background results were 119 ± 2 for L6 lot samples (L6A1-L6A6) and 122 ± 3 decays per minute for L8 lot samples (L8A1-L8A6).

Table I: Activity concentration of ^{36}Cl analyzed by LSC in nuclear waste samples. Results are expressed as average and Standard Deviation (SD) of triplicate measurements.

^{36}Cl activity concentration (Bq g^{-1})		
Sample aliquot	Average and SD ($n = 3$)	DL (3-sigma from BG)
L6A1	7.8 ± 1.8	2.7
L6A2	43.8 ± 2.1	3.1
L6A3	67.2 ± 1.7	2.9
L6A4	6.4 ± 1.1	2.9
L6A5	50.3 ± 1.4	3.3
L6A6	58.4 ± 1.9	1.8
L8A1	87.9 ± 2.4	3.7
L8A2	15.3 ± 2.1	2.8
L8A3	942 ± 5	8.3
L8A4	1.5 ± 1.2	2.8
L8A5	14.8 ± 2.3	3.0
L8A6	8.0 ± 0.92	2.0

4. Conclusions

Chlorine-36 activity concentrations were above DLs for all samples analyzed, indicating that the applied methodology is suitable to determine ^{36}Cl activity concentrations in nuclear waste samples from nuclear centers. This is a preliminary study and it is necessary to analyze a larger number of samples for more information.

Acknowledgements

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