COMPARISON OF ^{210}PB DETERMINATION IN ENVIRONMENTAL SAMPLES BY LIQUID SCINTILLATION COUNTING AND GAS FLOW PROPORTIONAL COUNTING

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Abstract. *In this paper, the two most routinely used analytical techniques for Pb determination in environmental samples, Liquid Scintillation Counting and Gas Flow Proportional Counting, are reviewed; with emphasis on radionuclide separation and purification, source preparation and measurement techniques. The accuracy, precision, selectivity, applicability and minimum detectable activity of the two techniques are presented.*

Key words*: Liquid Scintillation Counting, Gas Flow Proportional Counting, lead 210*

Introduction: Pb-210 is important from the viewpoints of radiation protection and environmental protection, due to its high toxicity. It is part of the U-238 natural series, formed by the decay of the gas Rn-222, with a half-live of 22.3 years and emission of beta particles with 16.5 keV (80.2%) and 63.0 keV (19.8%). This radionuclide can be determined by many techniques, such as Liquid Scintillation Counting (LSC) and Gas Flow Proportional Counting (GFPC). The LSC is a technique in which the sample is mixed to the liquid scintillation, forming a scintillation solution, capable of converting the kinetic energy of nuclear emissions into light photons. The interactions of the emissions occur in the liquid solution, producing excitement with emission of photons of ultraviolet radiation. The GFPC consists of an anode, mylar window and a gas flow P-10 (mixture of argon and methane). Alpha and beta particles penetrate the window and ionize the gas, resulting in further ionizations. These ionizations are accumulated at the anode to produce electrical pulses, and alpha and beta measurement are separated based on rise time and pulse height discrimination. This paper reviews the two techniques in terms of accuracy, precision, selectivity, applicability and minimum detectable activity (MDA) for Pb-210 determination in environmental samples

Materials and Methods: The verification of the two methodologies studied was performed using reference materials IAEA-326 – Radionuclides in soil, IAEA-385 Irish Sea Sediment and participating in Proficiency Tests (PT) organized by Instituto de Radioproteção e Dosimetria (IRD/CNEN).

The determination of Pb-210 by LSC measurement was performed using a 1220 Quantulus™ Ultra Low Level Liquid Scintillation Spectrometer. The samples containing the standard reference materials were mixed with Pb^{+2} carrier solution (30 mg mL^{-1}) and were percolated through a glass column filled with 3 g of Sr-Spec resin from EICHRON, pre-conditioned in HCl 2M. In this condition, the Pb-210 and Bi-210 are retained. A volume of 80 mL of HCl 2M was used to elute the interfering element Bi-210. In the next step, Pb-210 was eluted with HCl 6M. The solution was dried and the residue dissolved in a hot solution of 20 mL HNO_3, and 0.4g of oxalic acid was added. The pH was adjusted with NH_4OH and the Pb-210 was precipitated as a Pb oxalate and filtered in a Whatmam 40 filter. The chemical yield was determined gravimetrically. The filter was transferred to a vial with 15 mL of scintillation solution Hisafe III and the measurement was performed using a Quantulus. The chemical yield achieved varied from 42 to 75 %. The MDA achieved was 6 mBq per sample for a counting time of 24,000 s.
The Pb-210 determination by GFPC measurement was performed using a low background gas flow proportional detector (10-channel Low-Level Planchet Counter LB 770 Berthold). The samples containing the standard reference materials were mixed with Pb^{+2} carrier (20 mg mL^{-1}). Pb-210 concentration was determined by radiochemical procedure that consists of an initial precipitation of Pb with 3M H_2SO_4, dissolution of the precipitate with nitrilo-tri-acetic acid at basic pH, and precipitation of $^{210}PbCrO_4$ with 30% sodium chromate. The Pb-210 concentration was determined through its decay product, Bi-210 by measuring the gross beta activity of the $^{210}PbCrO_4$ precipitate. The chemical yields were determined by gravimetric analysis, the results obtained varied from

80% to 93%. The MDA achieved was 4 mBq per sample for a counting time of 7,200 s.

Results and Discussion: The results obtained for the concentration of Pb-210 by LSC and GFPC and the corresponding precision and accuracy are presented in Table 1 and Table 2, respectively.

Table 1 Concentration of Pb-210 by LSC and GFPC

Reference materials	Reference value	mean value LSC	mean value GFPC
PT/IRD August/11	1.43±0.29	1.40±0.1	1.56±0.08
PT/IRD December/11	2.80±0.6	2.71±0.05	2.50±0.08
PT/IRD April/11	0.50±0.10	0.46±0.05	0.45±0.03
IAEA - 326	53.3	56.1±3.8	56.8±3.2
IAEA - 385	32.9	33.6±3.5	34.7±2.5

Table 2 Precision and accuracy for the determination of Pb-210 by LSC and GFPC

Reference Materials	Relative Error % (LSC)	Relative Standard Deviation (LSC)	Relative Error % (GFPC)	Relative Standard Deviation (GFPC)
PT/IRD August/11	2.1	7.1	9.1	5.1
PT/IRD December/11	3.2	1.8	10.7	3.2
PT/IRD April/11	8.0	10.9	10.0	6.7
IAEA - 326	5.3	6.8	6.6	5.6
IAEA - 385	2.1	10.4	5.4	7.2

The relative error and relative standard deviation obtained are below 10%, giving evidence of the performance of the two techniques in terms of accuracy and precision.
The results obtained for the chemical yield for the methodology GFPC was better than LSC.
The two techniques presented similar sensibility, with MDAs of the same order of magnitude. However, for the LSC technique a counting time 3 times higher was necessary to achieve the same sensibility.
Comparing the two techniques in terms of selectivity, LSC separate and concentrate the radionuclides of interest by using a specific resin (Srspec); the GFPC technique use an extensive radiochemical procedure for the separation and concentration of Pb-210, which includes a series of precipitations and re-precipitations. Therefore, this technique is more expensive, more time consuming and use many chemical reagents, which final disposal is an environmental concern.
Another advantage of the LSC technique is that the spectra obtained allows discriminating the Pb-210 peak from other possible beta emitters interfering elements.

The figure 1 shows a typical spectrum of Pb-210 obtained by the LSC Quantulus equipment, with the region of interest adjusted manually.

Figure 1 Spectrum of Pb-210 using LSC

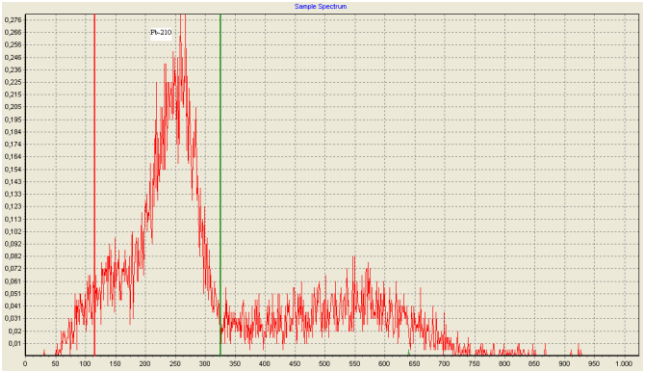

Fig. 1 Pb-210 spectrum of the LSC.

Concluding, the two methods described in this study are equally suitable for the determination of lead-210 in environmental samples.

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