

A FAST AND VERY SENSITIVE LSC PROCEDURE TO DETERMINE Pb-210 IN ENVIRONMENTAL SAMPLES

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Introduction

Pb-210, which has a half-life of 22,3 [1] years, as a member of the Uranium-Radium decay chain causes a significant contribution to the internal radiation dose and belongs with other members of this chain to the natural occurring radionuclides which show a very high dose factor for incorporation. But unfortunately this nuclide cannot be measured sensitive and fast enough in natural samples because of its low γ - ($E = 46,539$ keV [2], also low transformation yield of about 4 % [2]!) and β - energies (max. energies 17 keV and 61 keV [3] respectively).

This paper describes a method with both aims: lowest detection limit and short waiting time for the results. To reach this, after a radiochemical cleaning Pb-210 is detected in a liquid scintillation counter by its beta decay.

Methods

After drying the sample material at 105° C plant samples were reduced to small pieces and ashed at 450° C. To remove the remaining carbon fractions HNO₃ must be added to the ashes as often as necessary.

Soil samples must be sifted so that only fractions smaller than 2 mm remain. For the further procedure 40 ml HNO₃ and 10 ml HF were added to samples up to 5 g ash mass and evaporated to dryness. This step must be repeated two times. The remaining material must be treated with 2 ml HClO₄ for three times and evaporated to dryness.

From now on plant and soil samples are treated uniformly. The next step in sample preparation is a radiochemical cleaning. Thereby the ash is dissolved under warming in 100 ml HBr and extracted with 75 ml Trioctylamine/Toluene. The organic phase must be washed with 0,1 M HBr. For the reextraction of the lead 50 ml HCl must be added and the watery phase is mixed with 50 ml CHCl₃. The organic phase is discarded. After adding 50 ml HNO₃ the solution is evaporated to dryness. At this point lead is completely separated from all other elements except bismuth. Therefore radiochemical cleaning must be continued [4].

For that purpose the evaporate is dissolved in 0,02 M HCl and put on an anion exchange resin (DOWEX 1X8) column (3,5 x 0,8 cm) with a velocity of 1 ml/min. The anion exchange resin must be in the chloride form. This method elutes lead, the bismuth fraction remains on the column. The eluted fluid is evaporated to dryness taken up in 10 ml of 0,1 M HNO₃ and added to 10 ml of a scintillation cocktail (Quickszint 400 from Zinsser).

The chemical yield for lead was determined for each sample by atomic absorption spectrometry (AAS).

To detect Pb-210 by its low energy beta radiation, we used a liquid scintillation counter Quantulus 1220 from the Finnish company LKB-Wallac with an active shield.

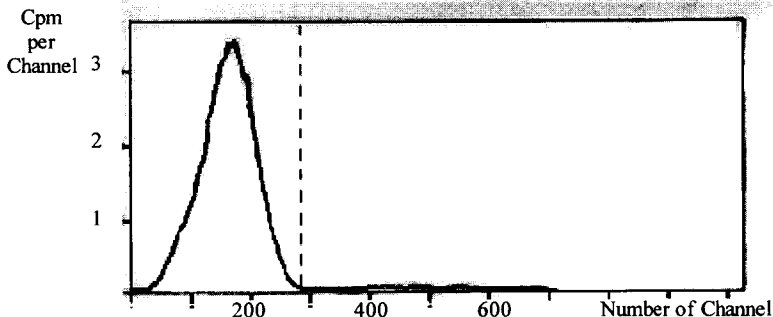


Figure 1: Spectra of Pb-210 with an activity of $(10,1 \pm 0,3)$ Bq Pb-210. The quenching parameter SQP(E) is 653. The counting window of Pb-210, which was fixed later is also drawn.

The time between separation of Pb-210 and start of the measurement was 3 h 55 min. In this period there is a build-up of Bi-210 and therefore the counts from channel 300 to 600 only come from this nuclide. Shorter waiting times are not desirable because of the effect of the chemoluminescence.

Fixation of the counting window for Pb-210 and the effect of quenching

In figure 1 the window for Pb-210 in the spectrum is marked. It reaches from channel 1 to an upper limit which is dependent to the effect of quenching. Quenching occurs when the sample is coloured, because the transmission of light pulses of the scintillator molecules to the photo cathode is weakened in the scintillator sample cocktail by absorption and so the spectra is shifted to lower energies. The quenching parameter SQP(E) which is detected by the LSC-Quantulus itself by a formula is a parameter for that effect. To get the dependence of the counting window for Pb-210 and the quenching parameter a preparation with different coloured samples was made. The result is a linear function between UC and SQP(E).

$$UC = a_1 \cdot SQP(E) + b_1 \quad (1)$$

with: UC: upper channel of the window of Pb-210

$$a_1 = 0,661$$

$$b_1 = -147,3$$

With each SQP(E) given by the LSC counter the suitable upper channel of the counting window can be calculated.

The role of Bi-210 and Po-210 in the spectrum

Immediately after separation, Bi-210 grows up again and disturbs the detection of Pb-210. Also Po-210 grows up, but this nuclide with its great half-life in comparison of the time after separation can be neglected.

To correct the spectrum of Pb-210 in the counting window from the counts which belong to Bi-210 it must be shown, that the ratio from counts of Bi-210 inside the window to those outside of it is constant with time.

After the measurement of a pure spectrum of Bi-210 this ratio could be fixed as the constant value of $0,2958 \pm 0,0009$.

When Pb-210 in a sample material is now detected the counts outside the detection window of Pb-210, which all belong to Bi-210 can be measured. With the constant ratio mentioned above it is possible to subtract this fraction from the counts in the detection window.

Counting efficiency for Pb-210

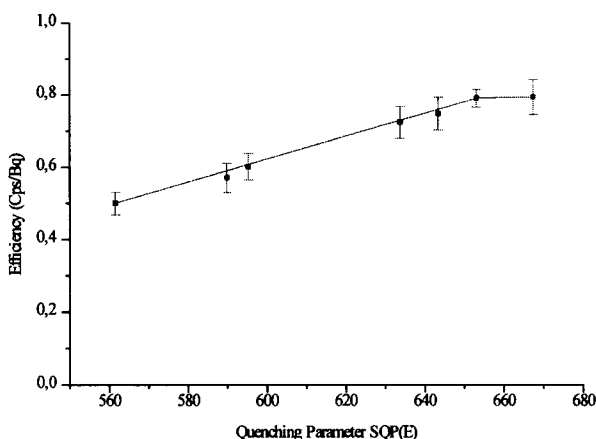


Figure 2: Relation of the counting efficiency for Pb-210 and the quenching parameter SQP(E) (1σ confidence level).

The quenching parameters normally are higher than 653, so that the counting efficiency is constant and $\eta = (0,80 \pm 0,04)$ Ips/Bq.

Influence of Pb-212 on the spectrum of Pb-210

Pb-212 from the thorium decay chain and its daughters cover a wide range of the measured spectrum by determination in LSC Quantulus. Pb-212 with a half-life of 10,64 h [3], Bi-212 ($T_{1/2} = 60,6$ min), Po-212 ($T_{1/2} = 305$ ns) and Tl-208 ($T_{1/2} = 3,07$ min) therefore also cause a counting rate in the window of Pb-210. But if the ratio of counts inside the window of Pb-210 belonging to Pb-212 and its daughters, to that outside the window is regarded, a constant value over time is obtained. This value, however, is only constant for times after separation which are greater than 4 hours. Then it amounts to $0,349 \pm 0,002$. This time is also sufficient to eliminate the possible presence of Pb-214 ($T_{1/2} = 26,8$ min)

With known ratios of counting rates from Bi-210 and Pb-212 (and daughters) in- and outside the window of Pb-210, chemical yield and efficiency for Pb-210 the activity of Pb-210 in the sample can be calculated.

Detection limits and comparison with other possibilities of the measurement of Pb-210

To compare the single methods, detection limits are calculated for a 1000 min measurement and Pb-210 in watery solution. The formulas for detection limit calculate the 3σ confidence level.

Table 1: Comparison of detection limits of the different detector systems for Pb-210 in watery solution; time of measurement: 1000 min, no Pb-212 in the sample material.

		Detection limit (mBq/sample)
high purity Ge detector	100 ml Dose (content 100 ml)	662
	LSC-vial (content 8 ml)	234
NaJ detector	LSC-vial (content 20 ml)	340
LSC Quantulus		7,4
gasfilled counter	detection via Bi-210	3,2
α -spectrometry	detection via Po-210	0,3

Table 2: Comparison of sample preparation time

method	sample preparation	duration of time in days for		
		radiochemical cleaning	time between end of analysis and begin of measurement	sum
high purity Ge det.	not ashed	0	0	0
	ashed	0	0	2
NaJ, ashed	2	0	0	2
gasfilled counter	2	2	5	9
LSC Quantulus	2	2	0	4
α -spectrometry	altogether ca. 120 days			

Taking into account also the detection limits mentioned in table 1, the LSC Quantulus has a very low detection limit and the time between drying of the sample and begin of detection for Pb-210 is short.

To compare the results of this method with a high purity Ge detector, soil samples have been measured and the results harmonise within the error bars.

Conclusions

A new method for the detection of Pb-210 has been presented. This nuclide is measured by an LSC Quantulus, which has an extremely low underground counting rate. Therefore very low detection limits can be reached. Pb-210 can be detected after a waiting time of only 4 hours after separation from the daughter product Bi-210. This time is necessary because of the chemoluminescence and the possible presence of disturbing Pb-212.

References

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