ALPHA ACTIVITY MEASUREMENT WITH LSC

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ABSTRACT

Recently, we showed that the alpha activity in liquid samples can be measured using a liquid scintillation analyzer without alpha/beta discrimination capability. The purpose of this work was to evaluate the performances of the method and to optimize the procedure of the sample preparation. A series of tests was performed to validate the procedure of alpha emitting radionuclides extraction in aqueous samples with Actinide Resin, especially regarding to the contact time required to extract all alpha nuclides. The main conclusions were that a minimum 18 hours stirring time is needed to achieve a percent recovery of the alpha nuclides grater than 90% and that the counting efficiency of alphas measurements with LSC is nearly 100%.

Key words: alpha LSC, *Actinide Resin*, gross alpha measurements

Introduction

Assessment of any release of radioactivity to the environment is important for the protection of public health, especially if the released activity can enter the food chain. Such assessment demands a rapid, reliable and practical measurement technique. It is known that low level gamma spectrometry is a suitable method for the assay of gamma emitting nuclides in environmental samples if a sufficient quantity of sample is collected. However, for the assay of alpha emitting nuclides the task is much more complicated than that for gamma emitting nuclides. Because of the possible absorption of alpha particles by the physical matrix, long chemical separations followed by electrodeposition are usually needed. In the event of accidental releases of radioactivity, it is essential for the radiation protection authorities to assess possible contamination. The methods mentioned above are obviously not satisfactory for the rapid screening of alpha emitting nuclides [1].

Recently, we showed that the alpha activity in liquid samples can be measured using a liquid scintillation analyzer without alpha/beta discrimination capability [2]. The used method was based on an *Eichrom Technologies Inc*. procedure [3], which combines an extraction technology based on a special resin *(Actinide Resin)* with liquid scintillation counting (LSC). Since in this case the samples are homogeneously dispersed in the scintillator medium, the problems of sample self-absorption are avoided and high detection efficiency can be obtained [4]. Another important advantage of LSC is its rapid and convenient sample preparation.

The purpose of this work was to test and evaluate the performances of the method and to validate the procedure of the sample preparation.

Method

Alpha Particle Detection by Liquid Scintillation Counting

The liquid scintillation detection process involves converting the kinetic energy of the alpha particle into light flashes. The intensity of the light emission and energy of these alpha particles are directly proportional to one another. Most alpha particles are emitted with kinetic energies in the range of 2-8 MeV. However, much of the energy is not directly converted to light but is dissipated through molecular ionization. This ionization, coupled with molecular damage from decomposition and free radical formation, gives rise to a short-lived process called "ionization quench". Because of these phenomena, a 5 MeV alpha particle will appear to have the energy of approximately 500 keV or about one tenth of its particle energy. Therefore, all alpha-emitting radionuclides are detected approximately in the range of 200-800 keV. This makes detection and quantization possible by conventional liquid scintillation counting. Alpha emitting radionuclides, when quantitated by LSC, generally produce a symmetrical peak around the energy of the alpha particle (1/10 its actual energy) [5].

The Procedure for Gross Alpha Radioactivity Determination in Water Samples

In [2] we showed how the analytical procedure *Gross Alpha Radioactivity in Water* from *Eichrom Technologies Inc.* was implemented in the Radiation Protection Laboratory from INR – Pitesti, emphasizing the modifications from the original procedure, especially regarding the counting mode.
Shortly, the procedure consists in extraction of alpha emitting radionuclides from a water sample by batch contact with *Actinide Resin* (100 - 150 µm particle size resin), manufactured by Eichrom Technologies. Usually, 0.5 g of resin is stirred with aliquots of 100 ml up to 500 ml water sample, after that the resin is collected and transferred to a scintillation vial and the alpha radioactivity is counted directly by LSC.

Regarding the counting mode, the original procedure recommends a Liquid Scintillation Analyzer (LSA) equipped with a Pulse Decay Analysis (PDA) circuit, which permits the discrimination of pulses produced by alpha and beta radiation (*alpha/beta LSC mode*). Because our LSA (a Packard Tri-Carb® 2100TR model) has not the PDA option, for alpha activity measurement we used the *alpha LSC mode* of this conventional LSA, *without alpha/beta discrimination capability*. The alpha LSC mode can be applied only if the sample preparation technique ensures the absence of beta/gamma impurities or, if this is not possible, the influence of impurities can be eliminated by appropriate spectrum analysis techniques. Theoretically, the measurement efficiency of alpha activity is nearly 100% in this counting mode, even with severe quenching [5, 6].

Regarding the sample preparation, in the Eichrom's procedure is mentioned that a long enough contact time between resin and water sample must be used to extract all alpha emitters (minimum of 4 hours, but 18 hours recommended).

Experimental

Two kinds of tests were performed to evaluate the performances of the method using our Tri-Carb® 2100TR LSA.

The first of them was made to demonstrate that a 100% counting efficiency is obtained for alpha activity measurement, even for low activity concentrations. Using well-defined quantities (gravimetrically determined) from ^{241}Am solutions with known activity concentration a series of experimental samples with different levels of alpha activity were prepared. It was intended to obtain experimental samples

containing four decreasing levels of activity (of about 5 Bq, 0.5 Bq, 0.125 Bq and 0.075 Bq), corresponding to activity concentration levels of approximately 10 Bq/L, 1 Bq/L, 0.25 Bq/L and 0.15 Bq/L respectively, for aliquots of 500 mL of water samples. The samples were prepared using 15 ml of UltimaGold AB liquid scintillation and PE scintillation vials of 20 ml. An appropriate blank sample was prepared, also.

The second kind of tests one took into account the validation of the extraction procedure of alpha emitting radionuclides in aqueous samples with Actinide Resin, principally regarding to the contact time required to extract all alpha nuclides. A series of experimental samples were prepared using a ^{241}Am solution with known activity concentration of 5.31 ± 5.5% Bq/g. All samples were prepared adding 1 ml from this solution to an aliquot of 500 ml of demineralized water acidified to pH 2, adding 0.5 g of *Actinide resin* and stirring for a certain time. The stirring time was varied between 1 and 23 hours. The collected resin was mixed with 15 ml of liquid scintillation UltimaGold AB into a PE scintillation vial of 20 ml. In the same way, but without adding the radioactive solution in the water sample, an appropriate blank sample was prepared. All samples were measured using alpha LSC mode and percent recovery variation function of stirring time was determined.

Results and discussion

Counting efficiency evaluation

Examples of spectra obtained using alpha LSC counting mode for some samples from the first kind of tests are presented in **Figure 1** for the first three levels of activity concentrations (with 100 minutes counting time) and in **Figure 2** for the lowest level of activity concentration (500 minutes counting time). All spectra were acquired on the whole energy range (0-2000 keV) of the LSA and an alpha window comprised between 500 and 920 channels number (energy range of 250 – 460 keV) was used for calculations.

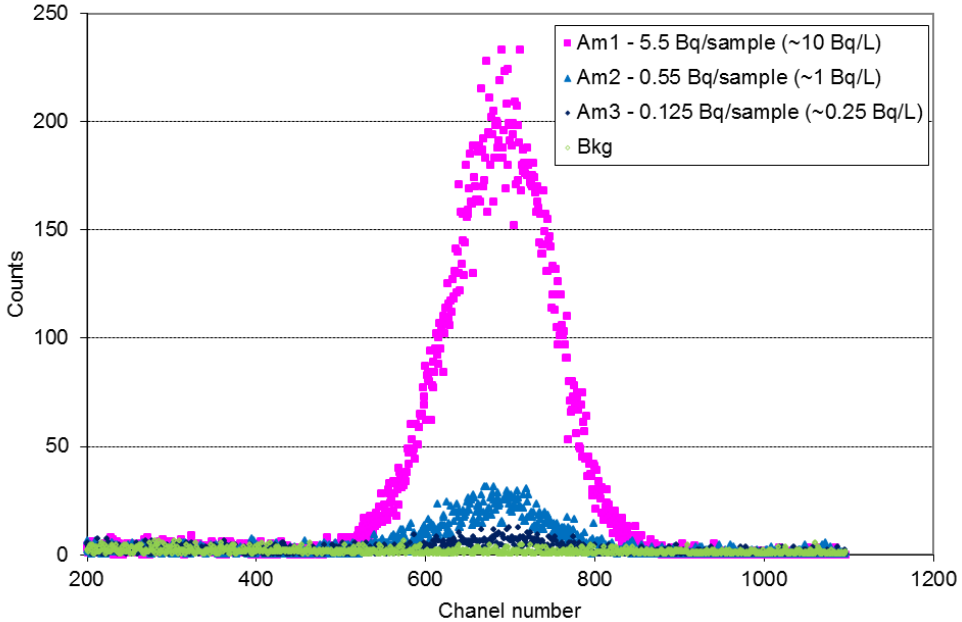

Figure 1 *Alpha LSC spectra (t_c = 100 min) for the first three levels of activity concentrations*

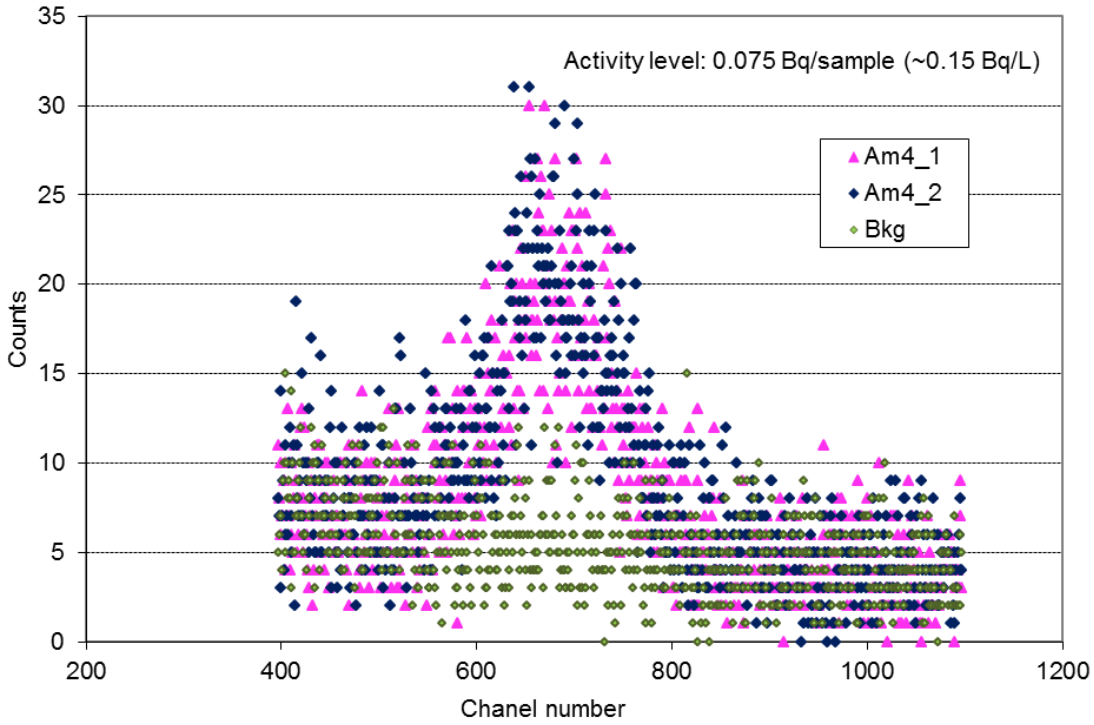

Figure 2 *Alpha LSC spectra (t_c = 500 min) for the lowest level of activity concentration*

The experimental results for measurement time of 100 minutes/sample are presented in the **Table 1**, as relative discrepancy between measured and known added activity for each sample, assuming a 100% counting efficiency. As can be seen, excepting for a sample having the lowest level of activity (which was only two times bigger than the background), relative discrepancies were lower than 3% for all samples, confirming that counting efficiency in alpha LSC mode is practically 100%.

Table 1 *Measured alpha activities of the experimental samples assuming a counting efficiency of 100%*

ID	Added (Bq/sample)	Measured (Bq/sample)	Discrepancy
aBkg	-	-	-
Am1_1	5.514	5.378	1.3%
Am1_2	5.496	5.423	2.2%
Am2_1	0.540	0.534	2.8%
Am2_2	0.538	0.533	2.5%
Am3_1	0.127	0.125	-1.7%
Am3_2	0.127	0.127	0.0%
Am4_1	0.073	0.073	0.3%
Am4_2	0.075	0.066	-11.6%

Percent recovery determination function of stirring time

Spectra of all experimental samples prepared as it showed above, obtained for a counting time of 60 minutes/sample, are presented in **Figures 3 – 5**, grouped according to the stirring time. Percent recovery (percent of measured activity divided to added activity) variation function of stirring time is presented in **Figure 6**.

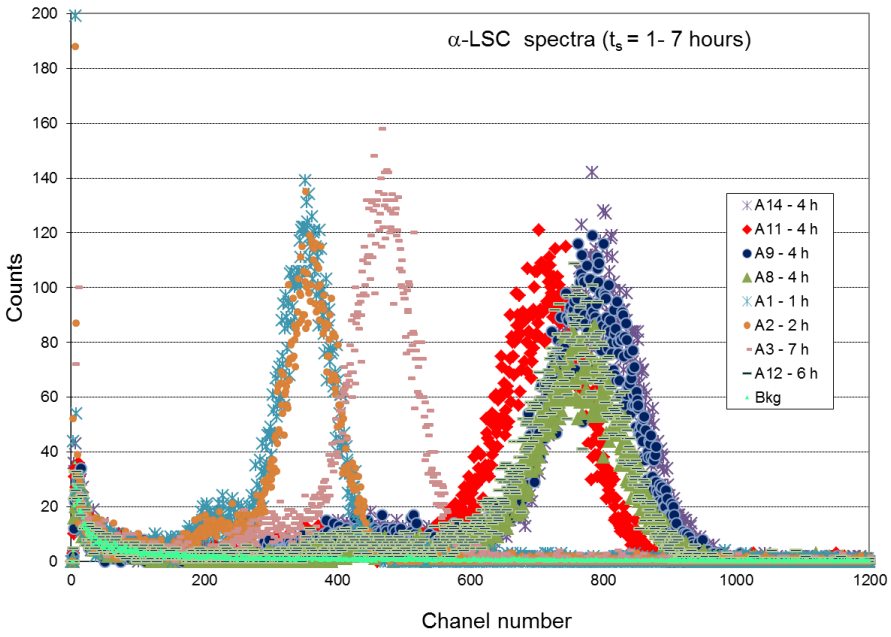

Figure 3 *Alpha LSC spectra for short stirring time (1 – 7 hours)*

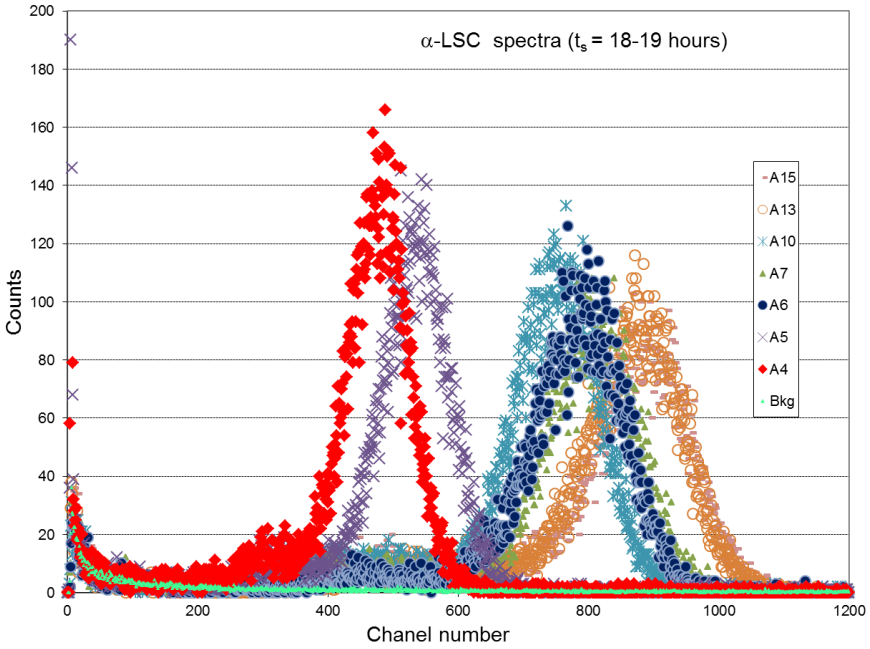

Figure 4 *Alpha LSC spectra for 18-19 hours stirring time*

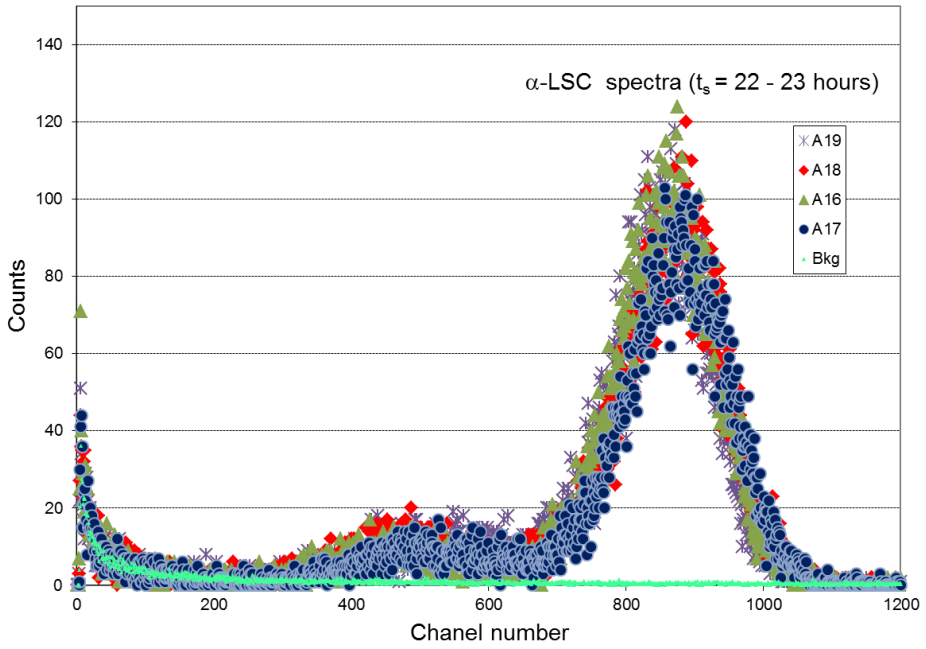

Figure 5 *Alpha LSC spectra for 22-23 hours stirring time.*

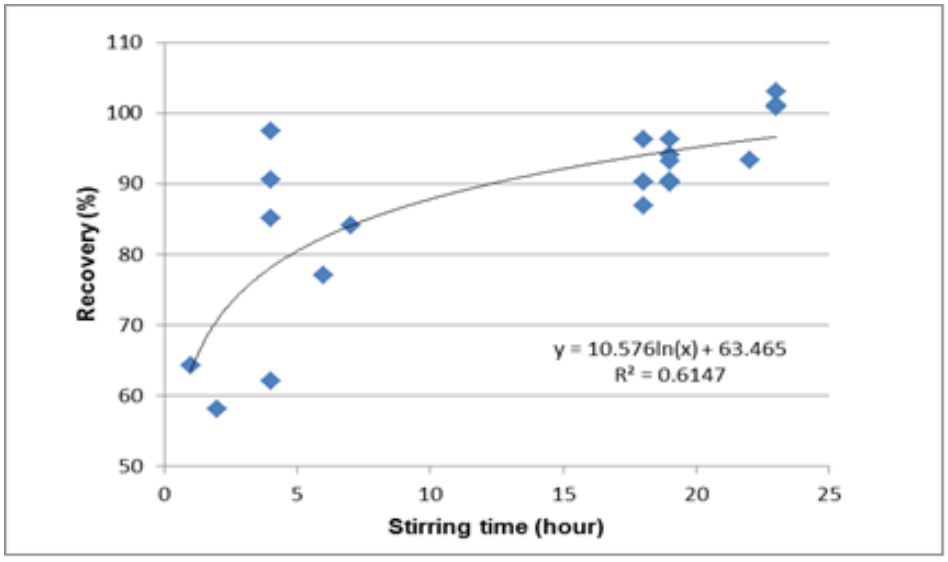

Figure 6 *Percent recovery function of stirring time.*

The analysis of the spectra showed that there are some differences, especially regarding the alpha peak position, which means that the extinction (chemical and/or color) had not a relatively constant level from sample to sample. The value of quench indicating parameter (tSIE) recorded of the LSA was, in each case, in accordance with the position of the alpha peak. However, different shape of alpha peaks in spectra is not an impediment, because counting efficiency in alpha LSC mode is virtually 100% irrespective of level of quench [5]. Parameter which proved to be very important in samples preparation was stirring time required resin to retain the best possible yield for alpha radioactivity in water samples. As can be seen in **Figure 6**, for a stirring time lower than 7 hours the percent recovery was generally

small, having scattered values. For stirring times of 18-19 hours the retaining performance was better, percent recovery being typically more than 90%, only in one case giving a value of 87%. For stirring times of 22-23 hours the percent recovery was practically 100%.

Conclusions

The method which combines extraction technology based on *Actinide Resin* with alpha LSC mode is the best choice when rapid screening for the presence of alpha activity in liquid samples is required, because alphas are counted with nearly 100% counting efficiency, even with severe quenching and for low levels of activity. A minimum 18 hours stirring time is needed to achieve a percent recovery of the alpha nuclides grater than 90%. The method may be used to quantitate alpha particles for a wide range of applications, such as environmental monitoring, nuclear fuel processing, high level waste management, etc.

References

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