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Determination of Actinides Using Digital Pulse Processing Analysis

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1. Introduction

The determination of low concentrations of actinides (Pu, U, Am, Np, etc) in environmental samples is vital for evaluating radioactive contamination caused by nuclear reactors, atomic bomb tests, and any type of nuclear incident. In an emergency situation, rapid analytical methods are essential to provide timely information to the authorities working to protect the environment and the population from the consequences of possible contamination.

Actinides disintegrate by emission of alpha particles which are difficult to detect because they are absorbed by the samples themselves. They are very dangerous to man because of the high relative biological effectiveness of alpha particles and their tendency to accumulate in several parts of the body (bones, kidney, liver, etc.) for many years because of their long biological and physical half-lives. Their determination usually demands lengthy analytical procedures which usually employ radiochemical pre-treatment of the sample followed by measurements using alpha spectrometry.

Alpha-emitting radioisotopes produce alpha particles at characteristic energies between 4 and 7 MeV, which can differ by as little as 10 keV, close to the smallest resolution of the silicon detectors used in alpha spectrometers. Alpha particles are heavy charged particles. Therefore, any physical medium between the alpha-emitting radionuclide and the detector will strongly absorb most of the alpha particle energy. These attenuations are produced by the sample itself or by any material between the sample and the detector. To solve this problem two different analytical procedures are commonly applied. A separation and concentration step is necessary in both procedures to avoid possible energy interferences between radionuclides and to concentrate in a small volume a significant amount of radioactivity, making it easier to measure. In order to account for any loss of the sample during separation, a known quantity of a specific isotope or tracer is usually added. The tracer is an isotope of the element under study with similar chemical behaviour. The radionuclide and tracer behaviour during the chemical treatment is the same, assuming that the tracer is homogeneously mixed and brought into chemical equilibrium with the sample.

The first procedure applies radiochemical sample treatment in an aqueous medium to separate and concentrate the different actinides before their measurement using high resolution passivated implanted planar silicon (PIPS) detectors (Holm, 1984; Aggarwal et al., 1985). This procedure allows for the alpha energies of the nuclides present in the sample to be identified with almost no interference. Thus very low minimum detectable activities (MDA) can be reached, but this process does require long analysis times. In this work we summarize this procedure and present results obtained in our laboratory using PIPS technology.

The second procedure using liquid scintillation (LS) is accurate and reproducible and considerably faster than other methods (Basson and Steyn, 1954; McKliven and McDowell, 1984; McDowell, 1986; Abuzwrda et al., 1987; Miglio and Willis, 1988; Lucas, 1980). The radionuclides are in a solution of an organic scintillator and there is no risk of sample selfabsorption, giving a counting efficiency of nearly 100%. Recent improvements have been made using organic cocktails which act simultaneously as extractors and scintillators, enabling elements to be separated in the relatively short time of several hours. However, the identification of the different radioisotopes is not simple because the resolution is low. The latest developments have focused on obtaining higher energy resolution by optimizing the chemical separation techniques and the extracting cocktails to provide the maximum amount of light emission and reduce quenching (McDowell, 1986). The electronics associated with these techniques have also been improved to reduce electronic noise and discriminate between beta and alpha pulses (McDowell, 1986; Reboli et al., 2005). The technique called photon-electron rejecting alpha liquid scintillation extracts the actinides from an aqueous solution into an organic phase containing the extractant, an energy transfer reagent, and a light emitting fluor. The counting efficiency reached is nearly 100% and the energy resolution is 5% for 4 to 6 MeV alpha particles. Using analogue pulse shape discrimination (PSD), it is possible to reduce the background from photo-electrons produced by external gamma-rays and to eliminate interference from beta emitters. PSD electronically selects pulses produced by alpha particles based on the longer decay time of their light emission (30 nsec), which is due to their much higher linear energy transfer to the scintillation solution.

The LS technique offers the advantage of a short analysis time but the resolution and the possibility of reducing interferences is limited. The initial way of increasing energy resolution was to use scintillation cocktails that produced more photons and improved light collection by using mirrors and a liquid between the sample and the photocathode to reduce light reflection (Hanschke, 1972; McDowell, 1994). The use of solid state detectors has also been used to improve resolution. Avalanche photodiodes (APD) have a larger gain than common photodiodes and are more sensitive to the frequency of the scintillator due to their greater spectral response. The replacement of photomultiplier tubes (PMTs) by APDs was successful for gamma-ray scintillation detection with energy resolution improvements and more compact detectors. Large area avalanche photodiodes have been applied to alpha liquid scintillation spectroscopy (Reboli et al., 2005). The influence of several parameters on energy resolution was studied: temperature, bias voltage, nature of the scintillating cocktail, and geometry of the counting vial. The improvement in energy resolution was attributed to the higher quantum efficiency and the more uniform active area of the photocathode compared to PMTs. The silicon photodiodes have a larger spectral response than PMTs, making it easier to match the fluorescence spectra of the scintillators with the APD spectral

response. However, APDs have the typical limitations of photodiodes: a limited gain and a higher noise contribution.

In this review we describe the experimental work performed in our laboratory in applying digital cards to the analysis of actinides in environmental samples and we compare our methodology with the classical procedures described above. The first section shows some results obtained using PIPS detectors. The use of PSD with fast digital cards was applied to LS spectrometry. Finally, we studied the results obtained by applying coincident liquid scintillation/high resolution gamma spectrometry to reduce interference and increase the limits of detection.

2. High resolution alpha spectroscopy with passivated implanted planar silicon detectors

This procedure consists of the radiochemical separation of the radioisotopes of interest and their electro-deposition to form a few atomic layers in order to avoid the loss of alpha particle energy. Such attenuation can be produced by the sample itself or by any material between the sample and the detector. The result is a characteristic tail in the alpha peak (Fig.1). To reduce the size of the tail the samples are counted in a vacuum camera and the samples have to be as thin as possible to avoid self absorption. Electro-deposition, evaporation and precipitation are the most commonly used procedures to produce the thinnest samples possible. The sample is placed in front of the detector inside a vacuum camera and data are acquired for a preset period of time. Achieving the desired lower limit of detection requires very long times because of the low activities involved. Count times of one or two days are common, and so the total analysis time including sample preparation can be several days.

The proper preparation of the sample is an important step in achieving high quality results in alpha spectroscopy. Different methodologies have been published for specific applications (Glover, 1984; Greeman et al., 1990; Gomez et al., 1998; Sarin et al., 1990). There are three principal steps in the preparation of an alpha spectroscopy sample: preliminary treatment of the sample, chemical separation and preparation of the source to be measured. The preliminary treatment is performed to homogenize the sample and to prepare it for the chemical processing. Different procedures are used for solid samples (e.g., food, soils, plants), liquid samples (water, blood, urine, etc.), and absorbents of gaseous or liquid samples (air filters, wipe type samples, etc.). In this early step the tracer nuclides are added to the samples.

Chemical separation is used to isolate and concentrate the elements of interest. Techniques used for separation include co-precipitation, solvent extraction, ion exchange and extraction chromatography.

The last step is required to prepare a sample suitable for being measured. The source preparation is an important step in obtaining the maximum resolution with an alpha spectrometer. The techniques used must be able to produce a very thin and uniform deposit to minimize the energy attenuation of alpha particles. The sources must be stable, and free of liquid, solvent and acid residues that could damage the vacuum camera or the detector. There are three main procedures for source preparation: electro-deposition (Singh et al.,1979), in which case the source is supposed to have a few atomic layers of the alpha emitter and the attenuation is extremely small; evaporation from an aqueous or organic solvent (Talvmtie, 1972; Aggarwal et al., 1985), which is sometimes followed by flaming

under vacuum (Glover, 1984) to remove the organic material; and co-precipitation (Singh et al., 1979 and 1983).

Fig.1 shows an alpha spectrum of a solution containing Ra-226 taken with a Canberra A450-18AM PIPS detector, with 450mm² of active area, 18 keV of alpha resolution, and a background of 6 counts/day. The sample was obtained after the evaporation of a few microliter droplets of the radioactive solution on a metal disk. Despite the small amount of residue left, about 0.1 mg/cm², the attenuation was still significant as can be seen by the long tails of the alpha peaks. The peak energies of the alpha radionuclides could be easily determined by looking at the peak edge position but the large tails make their quantitative determination difficult.

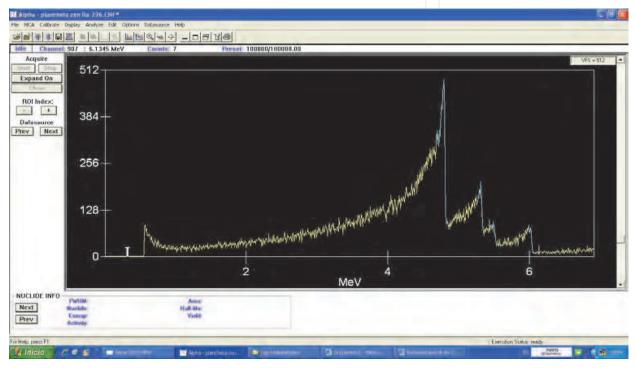


Fig. 1. Alpha spectrum of an evaporated solution of Ra-226 taken with a passivated implanted planar silicon detector. The software employed was Genie-2000, Canberra. The first peak situated to the left of the spectrum corresponds to Ra-226 (4.7 MeV), while subsequent peaks correspond to Po-210 (5.3MeV), Rn-222 (5.49MeV), and Pb-218 (6.0MeV).

Fig.2 shows the spectrum of a mineral sample containing U-238 and U-234. The sample was prepared by evaporation of the organic cocktail on a metal plate. The peaks of U-238 (4.2 MeV) and U-234 (4.7 MeV) are clearly visible but not the U-235 peak (4.5 MeV) that lies between both of them. The energy resolution was worse than in the case of aqueous evaporation because there was more organic residue (1 mg/cm²). The size of both peaks was the same because the sample contained natural uranium, and the U-238 and U-234 were approximately in radioactive equilibrium. The energy of the alpha peaks could be determined but a quantitative analysis was not possible because of the large peak tails. Chemicals to spread the sample solution can be added prior to its evaporation to obtain more uniform deposition, and the organic deposits burned off before counting, but this causes poor adherence of the sample to the backing and poor resolution. In some cases it is

possible to obtain a nearly solid-free deposit source sample. The procedure consists of evaporating the organic or aqueous sample to dryness, then treating with perchloric and nitric acids to oxidize the residual organic matter. The purified ion in aqueous solution is then extracted into thenoyltrifluoroacetone (TTA), deposited onto a stainless steel disc, and evaporated.

Sources are usually prepared by depositing micrograms of the element onto a flat, polished metal disk by electroplating from an aqueous medium. Properly prepared sources weigh less than $50~\mu g/cm^2$. In this case a resolution of 50-100~keV for 4-7~MeV alpha-particles can be reached. Fig.3 shows the alpha spectrum of a soil sample containing U-238, U-234 , U-235, and U-232 (the yield tracer). A small peak corresponding to U-235 can also be observed between the U-238 and U-234 peaks. The energy resolution of the spectrum enables all the uranium isotopes to be identified. Quantitative analysis can also be performed because the peaks are clearly defined and their area can be easily evaluated. The background is very low and it is possible to reach very small MDAs.

Cooprecipitation offers an alternative to electrodeposition which is too time consuming. The actinides can co-precipitate with small amounts of a rare-earth element carrier. The carrier elements may be precipitated as a fluoride by addition of hydrofluoric acid. The precipitate is filtered onto a 0.1 µm membrane filter which is dried, mounted onto a support backing, and used for alpha spectroscopy. The method is fast, inexpensive, and produces resolution nearly as good as electrodeposition.

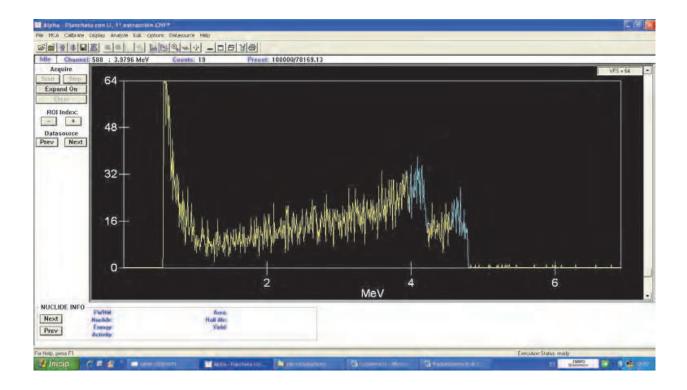


Fig. 2. Alpha spectrum of an evaporated organic extractant containing uranium. The U-238 (4.2 MeV) and U-234 (4.7 MeV) peaks (in blue) are clearly visible but not the U-235 peak (4.5 MeV) that lies between the two of them.

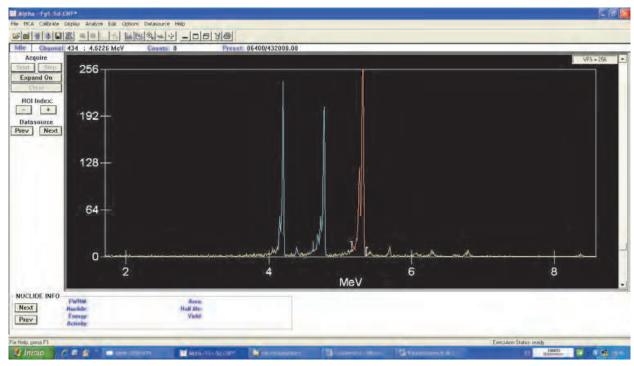


Fig. 3. Alpha spectrum of a soil sample containing U-238 (4.2 MeV), U-234 (4.7 MeV) (blue peaks), and U-232 (5.3 MeV) added as a yield tracer (red peak). A small peak corresponding to U-235 (4.4 MeV) can also be observed between the large peaks of U-238 and U-234.

3. Liquid scintillation alpha spectrometry using fast digital card and pulse shape discrimination analysis

The experimental equipment used in our laboratory to analyse actinides by LS spectrometry consisted of a standard photomultiplier tube with its pre-amplifier output directly connected to a fast digital card installed in a desktop computer. Two coincident photomultipliers were not needed, because the thermal electron noise in the phototubes was below the alpha pulse level. However, electronic noise, when superposed on the signal, could affect final resolution of the energy peaks. Usually, samples are immersed in a liquid to improve the transmission of light to the photomultiplier tube and they are surrounded by a light reflector to increase the efficiency of light collection. In our case the sample was placed directly above the photomultiplier tube. However, it has been observed that the geometry and reflector shape are important if good energy and pulse shape resolution are desired (McKlveen and McDowell, 1975).

The photomultiplier tube was a Saint Gobain RCA XP2412B with an AS07 preamplifier. It is possible to use several types of solid and liquid scintillators by placing them directly above the photomultiplier tube, but it is necessary to keep the phototube in a dark environment to avoid causing damage to the photocathode when a high voltage is applied. In our case the scintillator was the organic extraction cocktail containing the radionuclides of interest.

The typical analytical procedures to treat the solid samples were drying, grinding, and combustion of the sample to remove organic matter. Next, the sample was dissolved in a strong acid solution (nitric, perchloric and hydrofluoric acids). When samples contained silicate material, hydrofluoric acid was required, and repeated evaporation of the sample

dissolved in acid was needed to remove the silicates as silicon tetrafluoride. For the general case of samples without significant amounts of thorium or iron a two-stage extraction is a fast, simple separation procedure (Cadieux et al., 1994; McDowell, 19869). A first extraction from 0.8M nitric acid separated U and Pu into the organic phase. After the organic phase was removed, the aqueous phase was adjusted to a pH of 2.5 to 3.0 by the addition of a formate buffer solution. The second extraction is then performed to remove americium and curium. The extractive scintillator mixture contains 120g/l of HDEHP, 180g/l of scintillation grade naphthalene and 4.0g/l of 2-(4'-biphenylyl-6-phenyl-benzoxazole) [PBBO] dissolved in spectroscopic grade p-xylene. After the phases were separated the oxygen was purged to reduce quenching and improve energy resolution. Tests of the two-step extraction on water samples with known actinide activities have shown recoveries higher than 95% with a precision of 2% to 3% (Cadieux et al. 1994). In our case we found recoveries of 95% for the first extraction and about 80% for the second one with a precision of 5%.

PSD analysis is an essential part of the procedure and can be performed with analog or digital electronics. The recent availability of fast digital cards offers the advantage of greater reliability and simplicity of use. This is achieved because it removes the need for the electronic equipment necessary when employing conventional analog procedures, i.e. multichannel analysers (MCAs), amplifiers, coincidence electronics, single-channel analysers, etc. The direct analog-to-digital conversion of the detector signal and the use of software techniques allows to simplify tasks usually performed by the analog amplifier, i.e. electronic noise reduction, the treatment of pile-up pulses, trigger level restoration, and the reduction of ballistic deficit. MCAs are not needed because the digitized signal has all the necessary information to determine the pulse amplitude, which is proportional to the particle energy, and to obtain the energy spectrum. The direct analog-to-digital conversion of the detector signal offers many other possibilities (White and Miller, 1999; Warburton el al., 2000), such as alpha/beta/gamma discrimination by analysis of the electronic pulse shape (PSD), precise detection time determination of pulses by using time stamping information, the implementation of coincidence techniques, and dead-time reduction.

In order to test the accuracy of the procedure a standard liquid containing known activities of uranium (U-238, U-234, U-235), Am-241, and Sr-90 was employed for the analysis. The results from the first extraction, containing U-238, U-234, and U-235, and from the second extraction, containing Am-241, were place in two glass vials to be measured. The vials were situated directly in front of the photomultiplier tube to allow the maximum entrance of the light emitted by the sample. Other arrangements, adding reflectors to the vial or immersing it in liquid to improve light transmission, were also studied, but it was observed that different light paths produced a broadening of the energy peak and these procedures were abandoned.

The electronic signal at the pre-amplifier output was connected to a digital card (AlazarTechs ATS330 PCI) with the following technical specifications: two channels independently sampled at 12-bit resolution and a sampling rate of 50 million samples per second [Ms/sec], with multiple possibilities for triggering, multiple records and time stamping. The two channels are useful if coincidence measurements are performed with two independent electronic circuits and detectors. The device had been previously tested with different types of scintillation cocktails containing radioactive sources of known

radioactivity and different emitting radionuclides (Ra-226, Sr-90, Am-141), to establish their pulse shape characteristics, the optimum bias voltage, and the card parameters (sampling rate, samples per pulse, triggering thresholds, voltage range, input impedance, and noise level). Those hardware conditions determined the optimum parameters for particle detection when the organic liquid scintillator was coupled with the photomultiplier tube.

The digital card was supplied with basic software to set up the acquisition hardware. It contained a software development kit to allow full control of the card. In order to develop a complete system able to perform alpha/beta/gamma spectrometry and coincidence experiments, many other functions were developed and incorporated in the original software. The system developed acts like a digital oscilloscope that visualizes each individual pulse, and also like a conventional MCA. Several selection functions were incorporated, such as voltage amplitude, record length, sampling rate, trigger level, input impedance. The characteristics incorporated into the MCA were identification of a region of interest, peak area determination, calibration of energies, dead-time determination, and loading and recording of spectra.

Several algorithms were also developed to perform PSD analysis. They separated the signals that came from different scintillator types or were caused by different type of particles, allowing simultaneous acquisition of alpha/beta/gamma spectra from a radioactive sample. In addition, AND and OR relations could be used during acquisition to obtain a compound spectrum from the sample.

Fig. 4 shows the spectrum of a soil sample containing uranium. The spectrum was obtained with the organic cocktail from the first chemical extraction. It was difficult to differentiate the peaks for U-238 (4.2 MeV) and U-234 (4.7 MeV), because the resolution is 10% (0.5 MeV), larger than the resolution (5%) reported by other researchers using specific instrumentation (McKlveen and McDowell, 1984; Cadieux, 1990) or employing avalanche photodiodes (Reboli et al., 2005). Different procedures were used to reduce electronic noise (i.e. elimination of noise harmonics by using fast Fourier transform), but the best energy resolution reached was 8%.

Fig. 5 shows the spectrum of a sample containing Ra-226. The peaks for Ra-226 (4.7 MeV) and its descendants, Rn-222 (5.49 MeV), Pb-218 (6.0 MeV), and Po-214 (7.69 MeV) interfered and formed a unique peak. The small peak to the right corresponded to Po-214 (7.69 MeV). The sample was immersed in an aqueous medium containing Packard alpha/beta Ultima-Gold scintillator and a few drops of Ultima-Gold F scintillator to improve the energy resolution which was 8%.

The counting efficiency was 80%. Efficiencies near 100% could probably be reached with better energy resolution and light collection. A disperse background of counts was observed to the left of the peaks. That was attributed to pulses of lower amplitude produced by more attenuated light paths. The background was measured with a sample of the pure cocktail extractor. Its value was 10-4 counts per minute [cpm] below the energy peak area. It could provide an MDA close to 10 Becquerels per kilogram [Bq/kg] in the case of a ten-gram sample and a measurement time of one day, assuming the absence of energy interference with other radionuclides. In any case, the results obtained in terms of energy resolution and MDA are very similar to those obtained by using a Packard TRICARB LSC 2900TR.

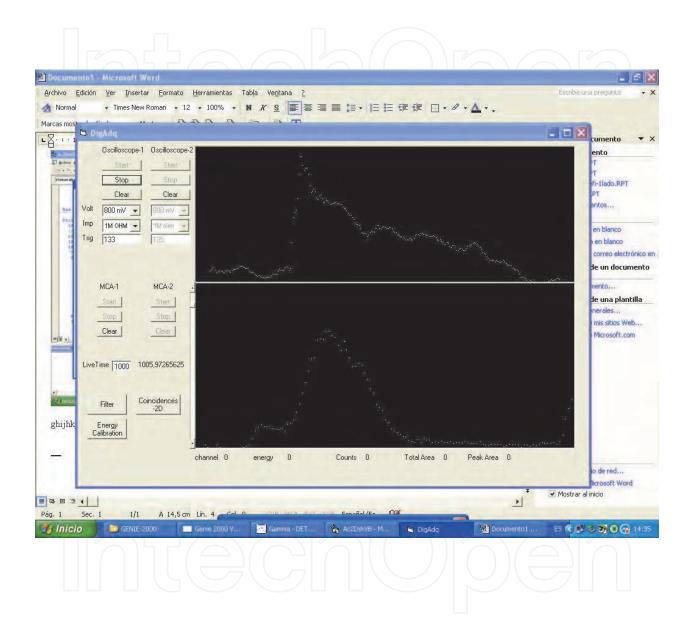


Fig. 4. Alpha spectrum of a uranium sample obtained by liquid-liquid extraction with organic solvents. The energy resolution was about 10% (0.5 MeV) and the peaks of U-238 (4.2 MeV) and U-234 (4.7 MeV) interfered. The upper part of the figure shows the shape of the electronic pulse at the preamplifier output. Each point of the pulse graph corresponds to 20 nsec.

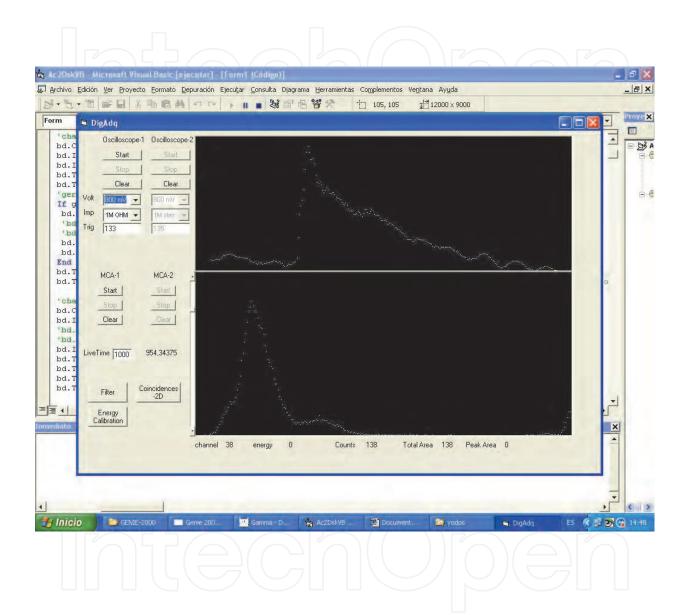


Fig. 5. Alpha spectrum of a radium sample in an aqueous medium using alpha/beta Ultima-Gold scintillator. The large peak corresponds to Ra-226 (4.7 MeV) and its descendents Po-210 (5.3 MeV), Rn-222 (5.49 MeV), and Pb-218 (6.0 MeV). They interfered and formed a unique peak. The small peak to the right is Po-214 (7.69 MeV). The upper part of the figure shows the shape of the electronic pulse at the preamplifier output. Each point of the pulse graph corresponds to 20 nsec.

4. Coincidence alpha X-ray in actinides using fast digital cards and pulse shape discrimination analysis

In order to increase the sensitivity of the procedure and reduce energy interference, gamma pulses emitted by actinides were measured in coincidence with alpha pulses. Many actinides decay by alpha particle emission in coincidence with low energy gamma/X-rays. When the alpha energies of two radionuclides interfere because their energy separation is below the energy resolution limit of LS spectroscopy, the coincidence procedure allows to obtain additional information by measuring the energy of the gamma/X-ray radiation coincident with the alpha emission.

The coincidence procedure is easy to implement by using a two channel digital card. The electronic procedure consists of recording on the second channel the X or gamma radiation coinciding with the alpha particles recorded on the first one. Using this method, it is possible to obtain X and gamma spectra with a significant reduction in background and the typical lead shield is not needed (de Celis, de la Fuente et al., 2007; de la Fuente, de Celis et al., 2008).

The detector employed for gamma radiation was a low-energy germanium, Canberra GL1515 with a 0.5mm Be window and a FWHM resolution of 304 eV at 5.9 keV and 551 eV at 122 keV. The detector area was 1500 square millimetres and its thickness 15 mm, which guarantees a high gamma-collection efficiency and a low energy resolution.

Tests were conducted with an Am-241 liquid source of known activity dissolved in 1ml of liquid scintillator. Am-241 emits 5.4 MeV alpha particles in coincidence with 59 keV gamma-rays. A plastic vial containing the radioactive source was placed directly over the photomultiplier tube and the germanium detector was situated above the sample. Small vials about 1cm in height were fabricated to reduce the distance between the liquid sample and the germanium detector. To implement the procedure in the digital card, the alpha trigger of the first channel is set "on" and the gamma trigger of the second channel is set to "off", in order to record gamma/X ray radiation only when the alpha channel is active. The pulses from the two detectors were stored jointly with the detection times and coincidences are identified by comparing the arrival time of the pulses proceeding from the liquid detector (alpha particles) and germanium detector (gamma/X rays). A delay time between pulses smaller than 1µs was considered to indicate a coincidence event.

Experimental tests were performed with a soil sample of known activity, previously reduced to ash and dissolved in nitric acid. The sample contained Am-241 and Pu-238 with activities below 1 Bq/kg. In conventional analysis most of these radionuclides are only detectable if radiochemical separation is combined with alpha spectrometry using PIPS detectors. With this procedure it was possible to identify and determine quantitatively the Am-241 activity. The MDA of the procedure was 0.01Bq/kg, which is very low compared with the 10 Bq/kg recorded when using directly gamma spectrometry and lower than the 0.1 Bq/kg achieved when using PIPS detectors. This is due to the high background reduction of the coincidence procedure. Other alpha/X-ray radionuclides, such as Pu-238, were not identified because of the efficiency reduction in the 10-20 keV region by the Ge absorption edge and X-ray self-absorption in the liquid sample. Tests to increase the MDA at these energies could be carried out with Si(PIN) detectors or using a phosphor sandwich [phoswich] detector with a higher resolution scintillator. Fig. 6 shows a coincidence event (alpha/gamma) and the coincidence

gamma spectrum of a soil sample. The gamma pulse was delayed with respect to the alpha pulse because an analog amplifier with a shaping constant of 1µs was employed to reduce electronic noise and obtain a better resolution in the germanium detector. Electronic noise reduction in the germanium chain could also be achieved using digital procedures.

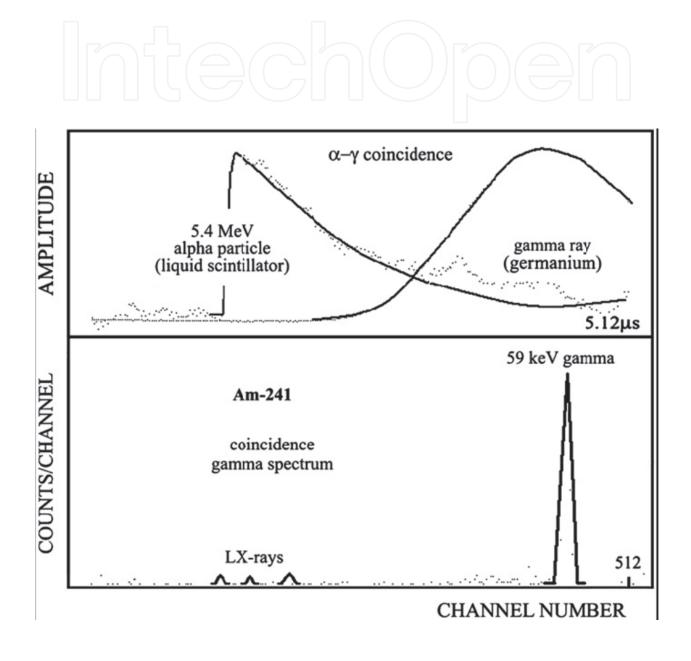


Fig. 6. Am-241 alpha/gamma coincident pulses (above) and coincident gamma spectrum (below) using liquid scintillation and a low energy germanium detector.

The procedure has the advantage of using the same geometry and chemical preparation for samples and standards, reducing the need for corrections for quenching. Interferences by beta/gamma coincidences are easily avoided, as beta particle pulses detected in the liquid scintillator are usually of smaller amplitude than alpha particle pulses. However, PSD, using the different scintillation decay time of alpha and beta particles, is intended for use in future. The use of a digital PSD system offers many other advantages, particularly the fact that the background of the detection system, which ultimately determines the sensitivity of the procedure, can be further reduced by studying the shape and detection time of the coincident particles, excluding those pulses which do not meet certain conditions.

5. Conclusions

Analytical procedures used to determine actinides in environmental samples were reviewed. The standard procedure using PIPS detectors is the most often used. It allows very small MDA to be reached due to the high resolution and very low backgrounds of these detectors. However, the analysis time is long (several days) and impossible to apply when many samples need to be processed or it is necessary to know the results in record time. LS spectrometry overcomes this inconvenience but at the expense of a poor resolution which makes it difficult to eliminate some interferences and thus determine quantitatively certain radionuclides. The LS analysis time is very short and improvements could be made to increase the light emission from the cocktail, reducing quenching and improving energy resolution. Future photodiode detectors could also help to improve energy resolution. A digital system is the natural way to implement many of these new developments due to its advantages in treating the digitized pulses. The MDA is ultimately dependent on the background of the technique. In our case it was possible to reduce the background by eliminating electronic signals which did not correspond with the correct pulse shape and also random electronic noise using standard digital procedures. Discrimination between different types of particles could be implemented by measuring the pulse light decay time without the need to resort to analogue electronic

Coincidence experiments could be easily set up using digital cards with two or more channels. The coincidence time could be determined with resolution times between 20 nsec and 1 nsec for cards of 50 Ms/sec to 1 Gs/sec sampling rates. Two-parameter studies using coincidence and 2-D diagrams can help to determine certain types of actinides. The technique is usually applied with analogue electronic equipment to determine radio-xenons resulting from nuclear subterranean tests but the setup presents more difficulties than when using digital equipment. In the case of actinide identification the technique could be applied to eliminate interferences between radioisotopes of similar alpha energies but different gamma emissions.

In some cases, for instance, the analysis of certain plutonium isotopes (Pu-239 and Pu-240), the alpha energies are similar and also equal the X-ray energy emissions. In this case conversion electron spectrometry with PIPs detectors using the same electro-deposited samples prepared for alpha spectrometry could help in obtaining a quantitative determination of both radioisotopes.

X-ray spectrometry could also be used to determine the total activity of the different radioisotopes of an element by using a high-resolution germanium detector in coincidence with LS spectrometry. The coincidence experiment is necessary to eliminate the background of the germanium detector and improve the MDA limit. Si (PIN) detectors, cooled by the Peltier effect, could offer a simple alternative if detectors of large area and small electronic noise are built in the future. Modern Si (PIN) detectors can reach energy resolutions of 125 eV at 5 keV with very low backgrounds but at the cost of a low counting efficiency because of their small detector area.

6. Acknowledgments

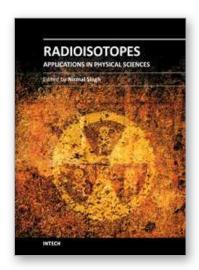
This research was supported by the Consejo de Seguridad Nuclear (CSN) of Spain, and the REM (Red de Estaciones de Muestreo) and PVRAIN (Plan de Vigilancia Radiológica Ambiental en la Central Nuclear de Sta. Mª Garoña) programmes.

7. References

- Abuzwrda M., Abouzreba S., Almedhem B., Zolotarev Yu.A. and Komarov N.A. (1987). Evaluation of a photoelectron-rejecting alpha liquid-scintillation, *Radioanal. Nucl. Chem.*, 111 (1987) 11.
- Aggarwal S.K., Chourasiya G., Duggal R.K., Singh C.P., Rawat A.S., and H.C. Jain. (1985). A comparative study of different methods of preparation of sources for alpha spectrometry of plutonium. *Nuclear Instruments and Methods in Physics Research*, Section A, Volume 238, Issues 2-3,1 August 1985, Pages 463-468.
- Cadieux, J.R. (1990). Evaluation of a photoelectron-rejecting alpha liquid-scintillation (PERALS) spectrometer for the measurement of alpha-emitting radionuclides. *Nuclear Instruments and Methods in Physics Research* Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, Volume 299, Issues 1-3, 20 December 1990, Pages 119-122.
- Cadieux, J.R., Clark S., Fjeld R.A., Reboul S. and Sowder A. (1994). Measurement of actinides in environmental samples by photon-electron rejecting alpha liquid scintillation. *Nuclear Instruments and Methods in Physics Research* Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, Volume 353, Issues 1-3, 30 December 1994, Pages 534-538.
- de Celis, B., de la Fuente, R., Williart, A., de Celis Alonso, B., (2006). Coincidence measurements in a/b/g spectrometry with phoswich detectors using digital pulse shape discrimination analysis. In: *ISRP10* Symposium, Coimbra, Portugal, 17-22 September 2006.
- de Celis, B., de la Fuente, R., Williart, A. and de Celis Alonso, B. (2007). Coincidence measurements in $\alpha/\beta/\gamma$ spectrometry with phoswich detectors using digital pulse shape discrimination analysis, *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, Volume 580, Issue 1, 21 September 2007, Pages 206-209. ISSN 0168-9002.
- de la Fuente R., de Celis B., del Canto V., Lumbreras J.M., de Celis Alonso B., Martín-Martín A. and Gutierrez-Villanueva, J.L. (2008). Low level radioactivity measurements

- with phoswich detectors using coincident techniques and digital pulse processing analysis *Journal of Environmental Radioactivity*, Volume 99, Issue 10, October 2008, Pages 1553-1557
- Glover, K. M. (1984). Alpha-particle spectrometry and its applications. *Int. J. Appl. Radiat. Isot.* 35, 239, 1984.
- Gomez Escobar, V., Vera Tome, F., Lozano and J.C., Martin Sanchez, A., (1998). Extractive procedure for uranium determination in water samples by liquid scintillation counting, *Appl. Radiat. Isot.* 49, 875-883.
- Greeman, D.J. and Rose, A.W. (1990). Form and behaviour of radium, uranium and thorium in central Pennsylvania soils derived from dolomite. *Geophys. Res. Lett.* 17, 833-836., 1990.
- Hanschke, T. (1972). High-resolution alpha spectroscopy with liquid scintillators by optimisation of the geometry, Ph.D. Thesis, Hannover Technical University, 1972.
- Holm E., (1984). Review of Alpha-Particle Spectrometric Measurements of Actinides, *Int. J Appl. Radiat . Isot .* 35 285.
- Lucas L.L. (1980). The Standardization of Alpha-Particle Sources, ASTM STP 698 (1980) 342. McDowell W.J. and McDowell L.B., *Liquid Scintillation Alpha Spectrometry*, CRC Press, Boca Raton, FL, USA, 1994.
- McDowell W.J., Nucl. Sci. Ser. on Radiochem. Tech. NAS-NS 3116 (1986).
- McDowell, W.J. (1980). Alpha liquid scintillation counting: Past, present, and future *The International Journal of Applied Radiation and Isotopes*, Volume 31, Issue 1, January 1980, page 23.
- McKliven, J.W. and McDowell, W.J. (1984). Liquid scintillation alpha spectrometry techniques, *Nuclear Instruments and Methods in Physics Research*, Volume 223, Issues 2-3, 15 June 1984, Pages 372-376.
- Miglio J.J. and Willis L.C., (1988). Simultaneous liquid scintillation determination of ²³⁹Pu and ²⁴¹Am in tissue, *J. Radioanal. Nucl. Chem.* 12 3 (1988) 517.
- Reboli, A., Aupiais, J. and Mialocq, J.C. (2005). Application of large area avalanche photodiodes for alpha liquid scintillation counting, *Nuclear Instruments and Methods in Physics Research* Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, Volume 550, Issue 3, 21 September 2005, Pages 593-602.
- Sarin, M.M., Krishnaswami, S., Somayajulu, B.L.K. and Moore, W.S. (1990). Chemistry of uranium, thorium and radium isotopes in the Ganga-Brahmaputra river system. *Geochim. Cosmochim. Acta* 5, 1990.
- Singh N.P., Ibraim S.H., Cohen N. and Wrenn M.E.,(1979). Determination of plutonium in sediments by solvent extraction and α-spectrometry, *Anal. Chem.* 50, 357 (1979) Pages 265-274.
- Singh N.P. and Wrenn M.E. (1983). Determination of alpha-emitting uranium isotopes in soft tissues by solvent extraction and alpha-spectrometry, *Talanta*, Volume 30, Issue 4, April 1983, Pages 271-274.
- Singh N.P., McDonald and Wrenn, E. (1988). Determinations of actinides in biological and environmental samples, *Science of The Total Environment*, Volume 70, March 1988, Pages 187-203.

- Talvitie N.A. (1972). Electrodeposition of actinides for alpha spectrometric determination. *Anal .Chem.* 44 (1972) 280-283.
- Warburton, W.K., Momayezi, M., Hubbard-Nelson, B., Skulski, W., (2000). Digital pulse processing: new possibilities in nuclear spectroscopy. *Applied Radiation and Isotopes* 53, 913-920. 2000.
- White, T. and Miller, W. (1999). A triple crystal phoswich detector with digital pulse shape discrimination for alpha/beta/gamma spectroscopy, *Nuclear Instruments and Methods in Physics Research* Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 422, 144-147.



Radioisotopes - Applications in Physical Sciences

Edited by Prof. Nirmal Singh

ISBN 978-953-307-510-5
Hard cover, 496 pages
Publisher InTech
Published online 19, October, 2011
Published in print edition October, 2011

The book Radioisotopes - Applications in Physical Sciences is divided into three sections namely: Radioisotopes and Some Physical Aspects, Radioisotopes in Environment and Radioisotopes in Power System Space Applications. Section I contains nine chapters on radioisotopes and production and their various applications in some physical and chemical processes. In Section II, ten chapters on the applications of radioisotopes in environment have been added. The interesting articles related to soil, water, environmental dosimetry/tracer and composition analyzer etc. are worth reading. Section III has three chapters on the use of radioisotopes in power systems which generate electrical power by converting heat released from the nuclear decay of radioactive isotopes. The system has to be flown in space for space exploration and radioisotopes can be a good alternative for heat-to-electrical energy conversion. The reader will very much benefit from the chapters presented in this section.

How to reference

In order to correctly reference this scholarly work, feel free to copy and paste the following:

B. de Celis, V. del Canto, R. de la Fuente, J.M. Lumbreras, J. Mundo and B. de Celis Alonso (2011). Determination of Actinides Using Digital Pulse Processing Analysis, Radioisotopes - Applications in Physical Sciences, Prof. Nirmal Singh (Ed.), ISBN: 978-953-307-510-5, InTech, Available from: http://www.intechopen.com/books/radioisotopes-applications-in-physical-sciences/determination-of-actinides-using-digital-pulse-processing-analysis



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