

Comparison of three measuring techniques performances to quantify Am-241 in different samples from laboratory experiments carried out under controlled conditions

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Abstract. Theoretically, alpha and gamma emission rates of Am-241 are intense enough to be actually used through three different radioactive measuring techniques, gamma spectrometry, liquid scintillation and alpha spectrometry. From an experimental point of view, it is very important to choose the less time-consuming method for measuring the isotopes activities from collected samples. Unfortunately, (for the metrologists) the behaviour of such an isotope as the Am-241 in a natural environment results in samples with very low incorporated activities. This is particularly true in terrestrial areas where transfers from soils to grown up plants are weak. Such a phenomenon leads to a wide range of sample contamination levels where, for example, soil activity can differ from plant activity by several orders of magnitude. Depending on what the radiological study is about, the metrologist has to apply the optimal measurement method regarding simultaneously, the sensitivity to be reached with the required sample preparation. In general, the lower is the sample activity, the more accurate the detection technique to discriminate between the effective signal from isotope under measurement and noise from natural ambient radioactivity must be. Spectrometric measurement techniques give, in this case, a quantitative result completed by qualitative information on reading the corresponding spectrum.

1. INTRODUCTION

The IPSN studies the transfer pathways and mechanisms for radioactive nuclides in the different components of natural ecosystems. The radioisotopes studied are usually present, in the environment, in the form of traces and originate initially from global fallout from atmospheric explosions following nuclear weapons testing. Locally these artificial radioisotopes may be present at slightly higher levels, although still very low, following controlled discharge from civil and military nuclear facilities. Amongst the artificial radioactive nuclides making up the discharge composition, the transuranians make up a group of elements, for which the high toxicity leads the LRE to study their behaviour in the controlled environment by means of experiments carried out in a laboratory. As a consequence, the laboratory must be able to perform measuring of these isotopes in the different matrices studied and at very different activity levels. This implies resorting several additional techniques chosen as a function of the level of contamination of the samples under consideration. The gamma spectrometry, liquid scintillation and alpha spectrometry are the three main radioactive measuring techniques used in this laboratory when the isotope to be measured possesses an emission spectrum (α, γ or β, γ), for which the different transitions feature sufficient intensity. The choice of one or another of these three measuring techniques is made mainly as a function of its sensitivity (detection limit), but also as a function of practical criteria such as preparation rapidity of samples for measuring. This procedure is illustrated through problems encountered during measurement of americium 241.

2. DECAY SCHEME OF AM241

2.1 α, γ Emission

The Am-241 disintegrates by giving off alpha particles. The main transition (85.2 %) leads to the emission of alpha particles having energy equal to 5,486 keV, leaving the daughter nucleus in an excited state. The unexciting is carried out by gamma transition, which leads either to direct emission of gamma photons of 59.6 keV, or to an electronic conversion followed by conversion electron emission. The gamma emission intensity is 35.9%.

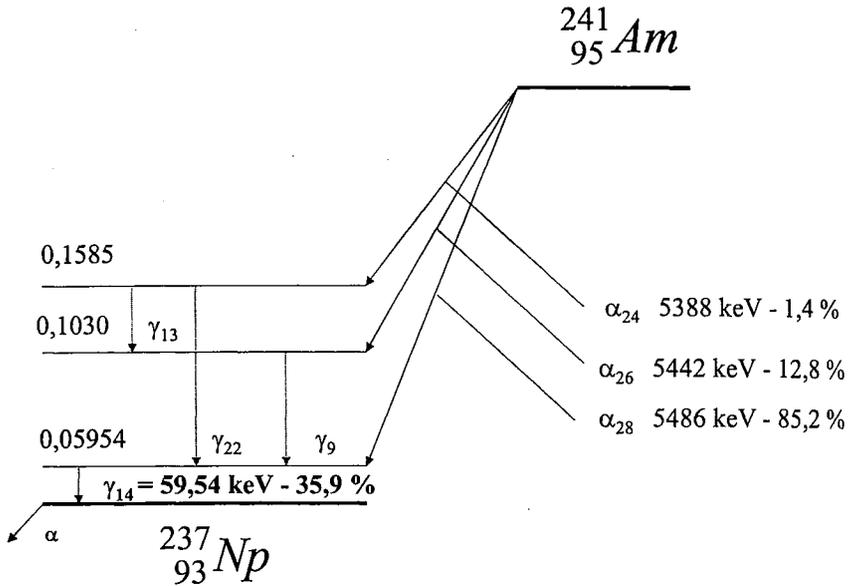
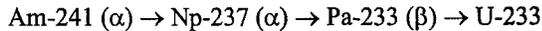


Figure 1: Main Am-241 emission.

2.2 Radioactive series

Am-241 is at the origin of a radioactive chain comprising a succession of radioisotopes before reaching stability with the ²⁰⁹Bi isotope. The first terms are as follows:



When working on a purified isotope, it is important to assess the incidence of the increase of descendants on the measurement of the parent isotope. Therefore, for an activity in Am-241 of 1 Bq at the time t = 0, one may observe that the descendants have an insignificant relative activity even after a very long time.

	Am-241 (433 years)	Np-237 (2,14.10 ⁶ year)	Pa-233 (27 days)	U-233 (1,6.10 ⁵ year)
t = 0	1	0	0	0
t = 1 year	0.998	3.10 ⁻⁷	0	0
t = 10 years	0.998	3.10 ⁻⁶	3.10 ⁻⁶	6.10 ⁻¹³

The calibrated solutions available on the market may therefore be stored for several years without requiring prior purification of the isotope at its descendants' level. There is no threat of any interference either by means of the "re-increase" of the daughter isotopes.

2.3 Measurement of 241-Am from experimental samples

Am241 emits α and γ radiations for which the rate of emissions enables it to be measured by different radioactive techniques. In the experimental field, the choice of one measuring technique will essentially be guided by the easiness and rapidity of its implementation, its ability to dose a large number of tests, its detection limit reached, its capacity to differentiate the researched signal from interfering signals. The Am241 study in the ground environment (soils and associated plants) leads to samples that present activity levels that may differ by several orders of magnitude. It is necessary to use several techniques by choosing the most appropriate for quantifying the isotope at the activity level under

consideration. In general, the lower the sample activity is, the more appropriate the measuring technique used must be to differentiate the researched signal from the ambient noise caused by natural radioactivity. The spectrometric techniques give, in this case, a quantitative response accompanied by qualitative information supplied by examining the obtained spectrum.

3. RADIOACTIVE MEASURING TECHNIQUES

The physical measuring techniques for radioactivity require the sample to be prepared beforehand in the optimised physical chemical shape for detection of the radiation under consideration α , β or γ . Such a sample is called the "source".

3.1 Gamma spectrometry

The gamma spectrometry is the technique offering the easiest sample preparation for measuring. The sample or representative aliquot of the latter, only needs to be prepared in a container adapted to the volume of the test piece, called "counting geometry" for which a detector efficiency calibration has been carried out. The detector (Hyper pure Germanium accompanied by its Dewar cryostat) is generally arranged in a measuring cell made up of a lead shielding intended to stop more or less completely parasite ambient radiations. A sample changer may be linked up to this cell enabling automatic change of the sample after programmed counting time. The control of this measuring cell is carried out by computer by means of a specific gamma spectrometry analysis program, which ensures both acquisition and processing functions of the spectrum obtained for each of the samples. Intrinsic characteristics of the detector, the shape and volume of the counting geometry, the thickness and quality of the lining, the energy and intensity of the emission peak used, the counting time applied, all depend on the detection limits reached for the isotope under consideration.

For a detector having a relative efficiency equal to 25% protected by a 10 cm thick lead shield, the detection limit obtained for Am-241 counted during 12 hours in a counting geometry of a volume equal to 50 ml is near to $5 \cdot 10^{-2}$ Bq. The minimal activity of the solution to be dosed should thus be higher than or equal to 1 Bq/l.

The apparatus equipped with a sample changer enables measuring 2 samples per day at this level of activity.

3.2 Liquid scintillation

Liquid scintillation is at the origin of the partiality technique used for the beta emitter measuring. For around ten years, the manufacturers have developed apparatus able to separate impulses originating from the emission of an β electron from those produced by a α particle¹. This same apparatus is also equipped with a guard circuit enveloping the measuring chamber, which enables coincidental impulses produced by the passage of a cosmic particle to be eliminated. The result of these two innovations is the possibility to dose alpha emitters with a very high sensitivity thanks to a very high detection efficiency (near to 100%) when coupled with very low background noise (near to 3 to 4 ipm).

In this technique, the detector is made of a scintillating mixture closely mixed with the aliquot of the sample to be dosed. One of the primary restrictions for using this technique is the proportions in which it is possible to associate both organic phases (scintillating mixture) and aqueous (sample) without considerably altering the detector mixture. In general, it is impossible to overshoot a 1/1 ratio, which leads to limiting the sample volume to ten ml at the most. For optimum use of this technique, it is advisable to use a low volume sample, in general 1 to 2 ml to which a scintillating mix additive would be added and be calculated in order to give a total volume equal to 20 ml. The procedure to follow will thus depend on the type of matrix to be dosed and the quantities of matter to be analysed.

For simple aqueous matrices, a quickly made pre-processing method consists of dry evaporating the aliquot and reusing this in a control volume (1 to 2 ml) of weak acid (HNO_3 , 0.1N). The aliquots of a batch of samples are thus introduced in the scintillating surroundings, leading to preparation of a series of samples with the physico-chemical properties that are relatively homogeneous.

For low volume solid matrices (plants, animal organs), samples should be dissolved and mineralised beforehand through action of an oxidising acid mix (such as concentrated $\text{HNO}_3 + \text{H}_2\text{O}_2$). The hot processing of samples enables them to be dissolved by organic molecule destruction. By carrying out some cycles of dry evaporation and reusing the 1ml concentrated $\text{HNO}_3 + 1 \text{ ml of } \text{H}_2\text{O}_2$, we may obtain a residue that is perfectly mineralised, it is then possible to dissolve, without any difficulty, in a minimal volume of diluted acid before introducing this in the scintillating cocktail.

For more refractory matrices and/or larger test pieces, there is, in general, no other possibility than to apply sophisticated radiochemical separation protocol enabling the researched element to be concentrated and purified. By completely eliminating the major elements making up the initial matrix, the different analytical sequences finally lead to a low volume column eluate or extract (organic or aqueous) being obtained that is easily incorporated into the scintillating mixture. However such separation protocols may not guarantee total recovery of the researched isotope. The analyst must then incorporate an internal tracer to the sample at the beginning of the analysis. This must be an isotope of the researched element, for which the initial known activity will enable him/her to calculate the chemical yield of the separation. By applying this correction factor to the value found for the researched isotope activity, the operator may accurately quantify the isotope real activity initially present in the test piece. This methodology implies that an isotope is available to play the role of internal tracer. In order to do this it must be distinguishable from the isotope to be dosed, either because of the nature of its emissions, or because of its energy is very different from an emission of the same nature.

As a function of the lining quality of the apparatus the detection limit is located between $2 \cdot 10^{-2}$ and 10^{-1} Bq. In the knowledge that the analysed sample is made of a maximum of 10 ml of radioactive solution added to 10 ml of scintillating mix, the solution to be dosed must show an activity higher than or equal to 2 Bq/l in order to be measurable by the most performing apparatus without the concentration stage.

Sample counting varies between 1 and 5 hours according to their activity. The apparatus equipped with a sample changer enables up to 24 samples/day to be measured.

3.3 Alpha spectrometry

Preparation of the source for counting, a crucial stage of any radioactive dosing, is of major importance here. This is due to the very low distance covered by alpha particles in the matter. The source should be made in such a manner as to avoid any self-absorption phenomena, which would imply a deposit being made "without thickness", that is completely transparent to the particle circuit from the emitter towards the detector. This is obtained by associating chemical separation protocols [2, 3], that are of very high performance, intended to eliminate all matter making up the initial matrix, with an electro-deposit technique that guarantees the homogeneous distribution of the researched element over the stainless steel disk surface. The different electro deposited element isotopes may then be quantified without difficulty considering the level of spectrometer resolution used.

In this process the radioactive element that is initially present in the form of traces scattered over the test piece is concentrated on an adjoining surface of around 1 cm^2 . By adjusting the test piece, the isotope quantity to be dosed may be adjusted so as to obtain significant counting. In environment samples of very low marking, this may lead to processing very large test pieces (up to 200g of soil). Obtaining such a result is, in this case, at the detriment of very demanding radiochemistry.

Alpha spectrometry is a spectroscopic technique that is particularly adapted to dosage of transuranians present in the form of traces in samples taken from the environment. Several chemical methods enable the main artificial elements to be sequentially separated and to dose the different isotopes. In the experimental field, the researcher is, in general, interested in the study of one particular element, studied through the isotope that is the easiest to quantify. The use of a spectrometric technique, does not seem to be *à priori* justified. However, study of transuranian elements in natural surroundings, shows the very low level of transfer of these elements, which leads to the production of samples that are little active. Finally, the use of proven separation techniques in conjunction with very low limits of detection allowed by the alpha spectrometry makes up an inevitable tool in several cases. Besides a very good spectrometer resolution, this technique is characterised by a very low background noise enabling the lowest detection limits to be reached.

The detection limit reached by this apparatus is near to 10^{-4} Bq for counting times equal to or higher than 1 week. When this activity is initially contained in a test piece of 10 g of matter (quantity widely available in experimentation), the corresponding sample activity is 10^{-2} Bq/kg. With a test piece of 100 g it is then possible to measure activities as low as 10^{-3} Bq/kg of matter.

4. CONCLUSION

Because gamma spectrometry generally requires a rapid and non-destructive sample preparation, it is particularly well adapted for measuring samples from experiments. The use of a sampler considerably increases the measuring capacity of a detector. Unfortunately the sensitivity of this technique is weak. Liquid scintillation can advantageously be used to measure large batch of samples after acid digestion. Because sample preparation can be slight and sensitivity is good, this technique is very well adapted to measure samples resulting of radioactive experiments. Alpha spectrometry remains the most sensitive technique and is well adapted for measuring environmental samples.

References

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