EPJ Web of Conferences **24**, 07004 (2012)
DOI: 10.1051/epjconf/20122407004

Optimization of $^{90}Sr/^{89}Sr$ measurements

M. Herranz*, R. Idoeta and F. Legarda

Department of Nuclear Engineering and Fluid Mechanics
University of the Basque Country (UPV/EHU)
Alameda de Urquijo s/n 48013 Bilbao, Spain

Abstract

One of the key points in the double measurement method for the measurement of both, ^{89}Sr and ^{90}Sr, by using a proportional counter is the choice of the times at which the measurements should be done. In this paper, the formulae to calculate the ^{89}Sr and ^{90}Sr detection limits in conditions of radioactive equilibrium between ^{90}Y and ^{90}Sr are derived, and an analysis of them as a function of the time between the two measurements is done. The choice for the time of the second measurement is going to depend on the desired quality of the results to be obtained.

1. Introduction

Among the fission products released by atmospheric nuclear explosions and nuclear power plants accidents to the environment, ^{90}Sr and ^{89}Sr are two of the most hazardous for man. In this sense ^{90}Sr and ^{89}Sr are two of the main radionuclides to be assessed by radiological monitoring procedures. It must be pointed out that the radiological toxicity of this element is relatively high as it has a tendency to be retained in bones, because its behavior is very similar to that of calcium.

^{90}Sr is also used in medical, research and industrial applications due to its half-life (29 y) and to the energy of its beta emissions (maximum: 546 keV).

*E-mail: m.herranz@ehu.es

After its radioactive decay the daughter nuclide ^{90}Y appears, which has a half-live of 64 h and a beta emission with end-point energy of 2280 keV. Again, the radioactive decay of ^{90}Y produces ^{90}Zr which is a stable nuclide. Due to this radioactive scheme, ^{90}Y grows in very quickly from ^{90}Sr after the release of strontium. In around 21 days radioactive secular equilibrium is achieved.

^{89}Sr is also a beta emitter with a half life of 50.53 days and the maximum energy of the beta particles that it produces is 1495.1 keV.

In the case of a release of radionuclides from a nuclear installation, at the time of fission, the ratio of activities from both activities ^{89}Sr/^{90}Sr is around 150 [1]. Only after a long time of the order of years, the activity of ^{89}Sr will be below detection limits and the samples will have only ^{90}Sr in secular equilibrium with its daughter ^{90}Y.

Any method to measure the radioactivity of Sr in a sample starts from the radiochemical isolation of this element from the sample, in order to separate it from any other beta emitters that could produce interferences in the measurement. Obviously, ^{89}Sr and ^{90}Sr nuclides are isolated together. In addition to this fact, at the instant that the Sr isolation process finishes, ^{90}Y grows in from the ^{90}Sr nuclide, producing counting-interferences in the measurement process of the strontium beta emitters.

This interference produced by ^{90}Y, ^{89}Sr and ^{90}Sr can be avoided by measuring the sample when secular equilibrium conditions between ^{90}Y and ^{90}Sr are reached. However, the interferences between ^{90}Sr and ^{89}Sr have to be born in mind and they can be quite relevant due to the relation of their activities in the sample.

Among the different methods to determine ^{89}Sr and ^{90}Sr [2], there exists a method that carries out two independent measurements of the same sample at two different time intervals [3, 4]. This is the method that is also employed in this study. In order to avoid the interference from ^{90}Y, the first measurement should be performed around 21 days after the strontium chemical separation process, that is, one has to wait until ^{90}Y and ^{90}Sr are in secular equilibrium. So, one of the key factors in this method is the choice of the time t_2, when the second measurement should be done. This choice depends on the required detection limits for ^{89}Sr and ^{90}Sr.

In this paper, starting from the formulae needed to calculate the ^{89}Sr and ^{90}Sr activities in the sample, and their respective detection limits in conditions of radioactive equilibrium between ^{90}Y and ^{90}Sr, an analysis is done about the detection limit that can be obtained for one strontium radioisotope in the presence of the other, as function of the time delay between the two measurements, $t_2 - t_1$.

2. Method

The product of the radiochemical isolation of the strontium from the sample is measured at two different times, t_1 and t_2, regarding the time in which strontium is isolated from Y as $t = 0$, thus, obtaining, respectively, two net counting rates, r_1 and r_2. The same counting time, T, is used in both, sample and background measurement.

The net counts rates, r_j, of these measurements can be calculated from the gross count rates, r_{jg} and the background counting rates, r_{j0}, as

$$r_j = r_{jg} - r_{j0},$$

being $j = 1, 2$ for the first and second measurement, respectively.

The net counting rates can also be calculated from the activities A_{90} from ^{90}Sr and A_{89} from ^{89}Sr in the sample at the time $t = 0$ by the following expression, taking into the radioactive equilibrium between ^{90}Y and ^{90}Sr:

$$r_j = 2 \cdot A_{90} \cdot \varepsilon_T + \varepsilon_{89} A_{89} e^{-\lambda_{89} t_j},$$

where ε_T is the counting efficiency of the simultaneous measurement of both, ^{90}Sr and ^{90}Y, ε_{89} the counting efficiency for ^{89}Sr, λ_{89} the decay constant for ^{89}Sr, and t_j the time of the measurement j since isolation.

From these expressions follows:

$$A_{90} = (r_2 - r_1 \cdot x)/(2\varepsilon_T(1 - x)), \tag{1}$$

$$A_{89} = ((r_1 - r_2)e^{+\lambda_{89} t_1})/(\varepsilon_{89}(1 - x)), \tag{2}$$

where $x = e^{-\lambda_{89}(t_2 - t_1)}$, is a parameter which considers the time delay, $t_2 - t_1$, between the two measurements. The time t_1 must be sufficient to achieve radioactive equilibrium between ^{90}Y and ^{90}Sr, *i.e.* at least 21 days.

The relative standard uncertainties of these activities, $u_{\text{rel}}(A_{90})$ and $u_{\text{rel}}(A_{89})$ can be calculated following the procedure reported in the GUM [5]. The relative standard uncertainties of the efficiencies have been considered, being $u_{\text{rel}}(\varepsilon_T)$ that of ε_T and $u_{\text{rel}}(\varepsilon_{89})$ that of the ^{89}Sr efficiency. Uncertainties of the decay constants and times have been considered as negligible:

$$u^2_{\text{rel}}(A_{90}) = ((r_{2g} + r_{20} + x^2 \cdot (r_{1g} + r_{10}))/(T \cdot (r_2 - r_1 x)^2)) + u^2_{\text{rel}}(\varepsilon_T), \tag{3a}$$

$$u^2_{\text{rel}}(A_{89}) = ((r_{2g} + r_{20} + r_{1g} + r_{10})/T(r_1 - r_2)^2) + u^2_{\text{rel}}(\varepsilon_{89}). \tag{3b}$$

The mathematical expressions for the decision thresholds and detection limits [6], are calculated according to the theory of Currie [7]. The decision thresholds, L_C^{90} for ^{90}Sr and L_C^{89} for ^{89}Sr, follow the expressions:

$$L_C^{90} = k_{1-\alpha} \cdot (1/2 \cdot \varepsilon_T \cdot (1-x)) \cdot \sqrt{(r_{20} + x^2 r_{10}) \cdot 2/T + x \cdot (x+1) A_{89} \cdot \varepsilon_{89}/T \cdot e^{\lambda_{89} t_1}}\,, \tag{4a}$$

$$L_C^{89} = k_{1-\alpha} \cdot e^{\lambda_{89} t_1} \varepsilon_{89}(x-1) \cdot \sqrt{(2(r_{10} + r_{20}) + 4 \cdot \varepsilon_T \cdot A_{90})/T}\,, \tag{4b}$$

where $k_{1-\alpha}$ is the quantile of the standard normal distribution for the probability $1-\alpha$.

The detection limits, L_D^{90} for ^{90}Sr and L_D^{89} for ^{89}Sr at $t = 0$, can also be calculated. According to Currie [7] for the detection limit can be used a different quantile, $k_{1-\beta}$, of the standard normal distribution for the probability $1-\beta$. But, as it is common in most laboratories, the quantiles $k_{1-\alpha}$ and $k_{1-\beta}$ employed in the decision threshold and in the detection limit, respectively, are assumed to be equal. In this case they are denoted as k, and the detection limits have the following expressions:

$$L_D^{90} = \frac{2 \cdot L_C^{90} + k^2 \cdot (1+x^2)/2 \cdot T \cdot \varepsilon_T (1-x)^2}{1 - k^2 \cdot u_{\mathrm{rel}}^2(\varepsilon_T)}\,, \tag{5a}$$

$$L_D^{89} = \frac{2 \cdot L_C^{89} + k^2 \cdot e^{\lambda_{89} t_1}(1+x)/\varepsilon_{89}(x-1)^2 T}{1 - k^2 \cdot u_{\mathrm{rel}}^2(\varepsilon_{89})}\,. \tag{5b}$$

At this moment it must be remembered that the unit for all these values is Bq, and refer to the activity in the sample.

In the two measurement method the different times, t_1 and t_2, in which the two independent measurements have to be carried out, must be chosen carefully, as this choice depends on the characteristics of the equipment used to measure the beta emissions and on the desired detection limits.

In this work, the equipment chosen was a gas flow proportional counter, provided with a 5 cm diameter and 0.5 μm thick Mylar window and with a typical background counting rate for beta counting of 0.8 min^{-1}. In this detector, ε_{89} has a value of 43% and ε_T is 40%, with standard uncertainties of 0.54% and 0.5%, respectively.

Following the radiochemical isolation of Sr from the sample a precipitate of Ba-Sr sulphate is obtained as a deposit on a cellulose-nitrate filter [3, 4], which is then measured in the proportional counter.

To carry out the analysis of the limits of detection, different time delays, $t_2 - t_1$, have been studied, more specifically, 7, 15, 21, 39 and 59 days. Also,

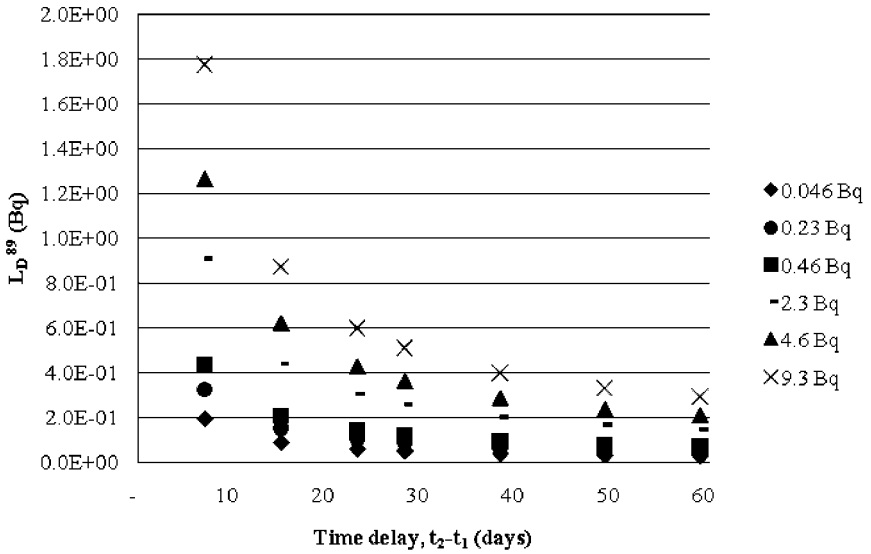

Figure 1: Detection limit of ^{89}Sr *versus* the time delay between the two measurements, for several values of the ^{90}Sr activity at the chemical separation date. The measurement time, T, chosen was 1000 minutes.

different ^{90}Sr and ^{89}Sr activities at the isolation date, ranging from cBq to daBq have been considered in this study.

3. Results and discussion

As shown in equation (5), detection limits depend not only on the uncertainties of the efficiencies considered and on the background counting rates, but also on the activity present in the sample from the Sr isotope whose detection limit is not the one considered, through the decision threshold, as well as on the time delay, $t_2 - t_1$, between the two measurements, hidden in the parameter x, and on the time t_1 when the first measurement is carried out.

From figure 1 it is clear that:

– The detection limit of ^{89}Sr increases with the ^{90}Sr activity. This behavior comes from the fact that the greater the counting rate from this last radionuclide is, the more difficult it is to determine the activity of ^{89}Sr.

– The detection limit of ^{89}Sr decreases as the time delay, $t_2 - t_1$, increases,

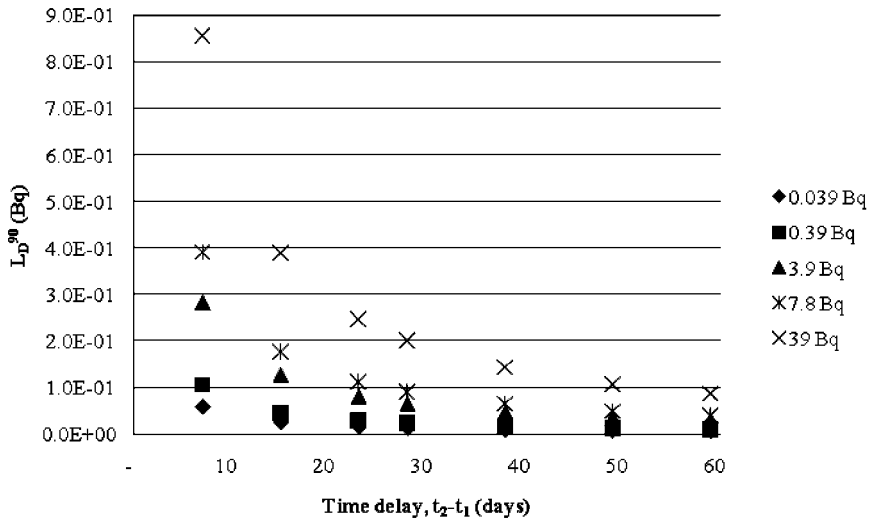

Figure 2: Detection limit of ^{90}Sr *versus* the time delay between the two measurements, for different values of the ^{89}Sr activity at the isolation date. The measurement time, T, chosen was 1000 minutes.

a trend which is independent of the activity ratio of the two strontium radioisotopes in the sample.

From figure 2 the following conclusions can be drawn:

– The detection limit of ^{90}Sr increases with the ^{89}Sr activity, as it is more difficult to determine the activity of ^{90}Sr, if a large counting rate from ^{89}Sr is present.

– The detection limit of ^{90}Sr decreases as the time delay between the two measurements, $t_2 - t_1$, increases, independently of the activity ratio in the sample and independently of the choice of t_1.

Thus, it can be said that the choice of the time delay between the two measurements depends on the detection limits required and also on the expected activity concentrations for both ^{90}Sr and ^{89}Sr.

If high activity concentration of ^{89}Sr is present in the sample it is going to be necessary to choose long time delays in order to observe a relatively low activity concentration of ^{90}Sr, slightly above the corresponding detection limit L_D^{90}. The same can be said, if one wants to observe relatively a low activity concentration of ^{89}Sr in presence of high activity concentration of ^{90}Sr in the same sample.

Obviously, as seen from equations (4) and (5), there are other parameters that can be varied to achieve lower detection limits: the larger the efficiencies are, the lower the decision thresholds and the detection limits become. This point cannot be modified to a large extent. The same can be said about the measurement time. The longer the time T is, the lower the decision thresholds and the detection limits are. Maximizing efficiency and counting time is a common task in all determinations in order to minimize the detection limits.

Considering a practical situation, the typical activity concentration found in drinking waters near in the north of Spain is about $5\,\mathrm{Bq\,m^{-3}}$. Thus, following figure 1, in order to observe an ^{89}Sr activity concentration of at least $0.6\,\mathrm{Bq\,m^{-3}}$ the second measurement should be carried out 15 days after the first measurement. But if the minimum detectable activity for ^{89}Sr has to be half that value, the second measurement should be done 38 days after the first one. This fact shows that this method has a major disadvantage: it is not rapid enough for situations in which a quick response about radiostrontium contamination has to be given.

4. Conclusions

For the simultaneous determination of ^{90}Sr and ^{89}Sr, the two measurement method with strontium separation and the use of gas proportional counters as radiation detectors is a helpful method. The method has been analyzed to establish before, which are the best conditions to carry out the measurement that guarantee that both radioisotopes can be determined.

The main conclusion is that longer time between the two measurements provides lower detection limits. Anyhow, the detection limit for one radionuclide is also influenced by the activity of the other strontium radionuclide, for example, if ^{90}Sr has a high activity, the detection limit for ^{89}Sr becomes high. The same is true vice versa.

References

[1] UNSCEAR report 1982, *Ionizing radiation: sources and biological effects* (United Nations, New York) 1982.

[2] VAJDA, N. and KIM, C., *Appl. Rad. Iso.*, **68** (2010) 2306.

[3] EPA 520/5-84-006 (1984) *Radiochemistry Procedures Manual: Method Sr-04* (EPA, Montgomery, USA) 1984.

[4] CIEMAT, *Procedimiento para la determinación de Sr-89 y Sr-90 en suelos y sedimentos*, Procedimiento específico Nº MA/07, Madrid, 1993.

[5] ISO, *Guide to the expression of uncertainty in measurement* (ISO, Géneve) 1993.

[6] CALMET D., HERRANZ M. and IDOETA R., *J. Radioanal. Nucl. Chem.*, **276** (2008) 299.

[7] CURRIE L.A., *Analytical Chemistry*, **40** (1968) 586.