Nuclear Engineering and Technology 56 (2024) 4661–4670

Contents lists available at ScienceDirect

Nuclear Engineering and Technology

journal homepage: www.elsevier.com/locate/net

Original Article

Alpha spectrometry: Avoiding recoil contamination of solid state alpha detectors

C. Olondo *, R. Idoeta, M. Herranz

Department of Energy Engineering, University of the Basque Country (UPV/EHU), Bilbao, 48013, Spain

ARTICLE INFO

Keywords:
Alpha spectrometry
Recoil contamination
Cover
Film

ABSTRACT

Due to its low background, alpha spectrometry is capable of determining very low activity concentrations of alpha emitters in all types of samples, including environmental samples. This low background can be lost due to detector contamination caused by recoil nuclei, which can even render the detector useless. In this paper, the thickness of a thin film used to cover the source is calculated analytically. This film, capable of retaining these nuclei and avoiding this contamination, allows all the alpha particles emitted by the source to reach the detector. The energy loss of the alpha particles passing through it was also estimated in order to identify the peaks in the spectrum, including the unexpected ones. The validity of the calculations carried out and the effectiveness of the film in retaining the recoil nuclei were verified using thorium samples. Finally, the effect of applying a negative potential to the source on the retention of these nuclei was studied. The study was carried out using a PIPS detector under vacuum conditions (0.1 mm Hg) with the samples placed at a distance of 5 mm from the detector. The recoil suppression systems of some commonly used commercial instruments were also tested.

1. Introduction

The analysis of radionuclides by alpha spectrometry involves the prior preparation of the sample, including the radiochemical separation of the element to which the radionuclide to be analysed belongs, and its subsequent electrodeposition on a stainless steel planchet (alpha source), which is placed in front of the detector. Due to its low background, alpha spectrometry is a very useful method for the analysis of alpha-emitting radionuclides, especially for the measurement of environmental samples. However, this capability can be reduced if the detector is contaminated by recoil contamination. This contamination is the result of recoil nuclei: when an alpha decay occurs in the alpha source, not only alpha particles are emitted, but also recoil nuclei, the decay product. Depending on their kinetic energy, the recoil nuclei can reach the detector and, if they manage to implant themselves on the detector, they will contaminate it if they are also alpha-emitting radionuclides. Alpha emissions from these recoil nuclei will not only affect the suitability of the activity measurement by interfering with the signal of the radionuclide being analysed, but will also increase the detector background and thus the detection limit of the measurement. The importance of keeping the detectors free from contamination by recoil nuclei is that it guarantees the adequacy of the activity determination carried out in the detector, since in the peaks associated with the isotopes whose activity is to be determined, only the number of counts corresponding to their emission is recorded, since it is not affected by the recording of counts from these recoil nuclei. Similarly, maintaining a low background and therefore a low detection limit (LD) makes it possible to comply with the regulatory standards set by the competent authorities, both within the framework of the surveillance plans carried out around nuclear power plants and in their decommissioning plans, with regard to the determination of the activity concentration in environmental samples: a low LD makes it possible to detect trace levels of activity concentration, if any, and to ensure that, if activity is present, it is below the limit set by the competent authorities, which is always very low. In the same way, a low LD makes it possible to adequately determine the value of the natural radiological background in the environment of a nuclear installation, a value to which it is necessary to return once the installation has ceased its activity, and guarantees the safety of the detector when measuring samples with high activity, such as samples from the NORM industry or from nuclear power stations undergoing decommissioning, since direct measurement of this type of source without taking precautions to avoid this type of contamination would greatly increase the probability of the detector being effectively contaminated. The significance of such contamination depends largely

* Corresponding author.
E-mail address: kontxi.olondo@ehu.eus (C. Olondo).

https://doi.org/10.1016/j.net.2024.06.029
Received 2 October 2023; Received in revised form 2 May 2024; Accepted 18 June 2024
Available online 25 June 2024

on the half-lives of both the radionuclide of interest and its decay products. If the decay product is a short-lived nucleus (a few hours), this is not too serious as the contamination will disappear within an acceptable period of time, but if it is a medium-lived nucleus (more than a few weeks or months), the contamination will take an unacceptably long time to decay [1]. In the case of longer-lived alpha emitters that decay to isotopes that are also long-lived alpha emitters, they do not pose much of a problem in terms of recoil contamination because the activity produced by these decay products will be small over a reasonable counting time (typically less than 600000 s); however, it is always important to keep it as low as possible. Based on the experimental results, two methods have been commonly used to avoid recoil contamination: applying a negative polarisation to the base where the alpha source is placed in the detection system, together with an air cushion between the alpha source and the detector, and covering the alpha source with a thin film. In fact, based on these results, some commercial detection systems have introduced different systems to avoid this contamination: from applying 3 V to the base to applying 5 V together with an air cushion.

Sill and Olson [1] demonstrated experimentally that recoil contamination of Fr-221 could be significantly reduced by applying a negative potential of 6 V and providing an absorbing air layer of 12 μg/cm^2, available at a vacuum pressure of 5–6 mm Hg and placing the detector 1.28 cm from the source. Inn et al. [2], using a source of Th-229, confirmed the validity of the method they had used to prevent recoil contamination [3]: a thin film collodion (about 12 μg/cm^2 thick) combined with a negative potential of up to 10 V; they proposed this film as an alternative for those who did not have detector units equipped with recoil suppression systems; they also pointed out that this collodion film showed damage after several weeks of exposure. Gels et al. [4] also analysed the use of a collodion film used by Inn et al. [2]) to avoid recoil contamination; they used a film of 8–16 μg/cm^2 thickness covering a source of Th-228 and analysed its effect on resolution. Vainblat et al. [5] analysed the effects of Mylar film coatings (0.25 mg/cm^2, 0.53 mg/cm^2 and 0.85 mg/cm^2) at different distances (1.7 cm, 2.9 cm, 4.1 cm and 4.9 cm) on resolution, efficiency and detection limit using a calibration source of Am-241, *Cm*-244 and Pu-239; they finally selected a Mylar film of 0.25 mg/cm^2. Janda et al. [6] heated the source in muffle furnace up to 1200 °C instead of establishing an air thickness and a negative potential or a film coating. Cosma et al. [7] studied the effect of formvar films [8] on alpha spectrometry parameters. All these studies are based on experimental measurements, but none of them has theoretically analysed the characteristics of the film in such a way that its effect on the emitted alpha particles would be negligible and would completely stop the recoil nuclei for any of the radionuclides usually determined by this technique and also for the tracers used. This is the aim of this work.

This work investigates, both analytically and experimentally, the ability of a thin film cover on the source to prevent recoil contamination. The study was carried out under the same conditions in which our laboratory performs alpha spectrometry: in low vacuum conditions (less than 0.1 mm Hg) and close to the detector (0.5 cm). It should be noted that our laboratory is dedicated to the measurement of environmental samples that are expected to have very low activity. As a result, alpha spectrometry measurements are carried out giving priority to high efficiency at the expense of better resolution (at 0.5 cm, the efficiency of the detector is about three times that obtained at 4.5 cm, while the resolution may decrease from 25 keV to 3.5 keV). The film used to cover the source is a mixture of cyclohexanone and poly(vinyl chloride-*co*-vinyl acetate-*co*-vinyl alcohol) called VYNS.

The first step is to analytically determine the thickness of the film that must be placed over the alpha source to avoid recoil contamination while still allowing the emitted alpha particles to pass through and reach the detector. This calculation takes into account all the recoil nuclei and alpha particles emitted by all the radionuclides whose activity is likely to be determined by alpha spectrometry. After determining the thickness of the film to be used, the energy loss that this film will cause to the alpha particles passing through it was also calculated in order to evaluate the spectral shift and thus to be able to identify possible interferences that might be observed in them.

Experimental measurements were then carried out on six samples of medium-high activity of Th-228, Th-229, Th-230 and Th-232 to validate the theoretical results obtained and thus the ability of this film to prevent recoil contamination. The samples were chosen because these radioisotopes decay to products with medium and low half-lives. All the samples were measured in three different situations: a) without applying any system to prevent recoil contamination, b) after covering the source with the VYNS to prevent recoil contamination and c) without covering the source but using the recoil suppression system provided by the equipment used to carry out the measurements: the application of 3 V to the base of the alpha source. After studying the results obtained from these measurements, four different negative potentials (3 V, 6 V, 18 V and 39 V) were also applied to the source, in order to study the possibility of applying only a negative potential to avoid recoil contamination. All the spectra obtained were also analysed in order to know the effect of the film used on the energy resolution. Finally, the state of the film was checked, which shows that it is in good condition despite the passage of time (after almost a year). As mentioned above, all calculations were carried out under our laboratory conditions: a low vacuum (0.1 mm Hg) is created and the alpha source is placed at a distance of 0.5 cm from the detector. In addition, and under our laboratory measurement conditions, the ability to avoid recoil contamination of the recoil suppression system, consisting of the creation of an air cushion together with the application of 5 V, was verified.

This paper presents the results of both analytical and experimental studies.

2. Analytical calculation and materials

2.1. Analytical calculation

Prior to the experimental measurements, and for all the main radionuclides whose activity can be determined by alpha spectrometry, the range in which all the recoil nuclei and alpha particles emitted by these radionuclides can be present in the VYNs was calculated, as well as the energy loss suffered by these alpha particles during their passage through these VYNs. These calculations were carried out taking into account the need to know both the kinetic energy of the recoil nuclei and that of the emitted alpha particles.

These calculations were therefore been carried out not only for natural emitters - the actinium, neptunium, thorium and uranium series - but also for other artificial radioisotopes such as Po-209, Am-241, Cm-242, Cm-2343, Cm-244, Cm-245 and Am-243, Pu-239 + 240, Pu-238 and Pu-242, although these are long-lived radionuclides with long-lived progeny. As a result, the emission of alpha particles from 2 MeV to 10 MeV was analysed.

From these calculations, the amount of VYNS needed to cover the source is obtained.

2.1.1. Kinetic energy

The kinetic energy of the recoil nucleus (T_R) and the alpha particle (T_α) produced during an activity measurement of an alpha emitter are calculated as follows [9]:

$$T_R = \frac{4}{A}(Q_\alpha - E_i)[MeV] \quad (1)$$

$$T_\alpha = \frac{A-4}{A}(Q_\alpha - E_i)[MeV] \quad (2)$$

Where A is the mass number of the alpha emitter, Q_α is the alpha decay energy or the energy available when an alpha particle is emitted, and E_i is the excited energy state of the recoil nucleus, where i indicates its energy level (the values used for Q_α and E_i are those provided by

Nucleide Lara [10], which also gives the kinetic energy of alpha particles).

2.1.2. Range of alpha particles in the film

The mean range of the alpha particles, i.e. the thickness at which the number of alpha particles emitted falls to half of its initial value, of alpha particles in the film was analysed to determine the maximum film thickness that could be used to ensure that all the alpha particles emitted for each isotope analysed reach the detector, so that the use of this film does not reduce the number of counts recorded.

The mean range in the film can be calculated from the mean range of the same particles in air at normal temperature and pressure, whose values can be obtained by using semi-empirical equations (3) and (4) [10]. The mean range should be about 5 % less, or even less, than the extrapolated range or thickness required to stop all the alpha particles emitted [10]. In this paper, when the term range is used, unless otherwise stated, it is the mean range.

$$R_{\alpha,air}\left[mm\right] = \exp\left[1.61\sqrt{T_\alpha}\right] \quad 1 < T_\alpha \leq 4\ MeV \tag{3}$$

$$R_{\alpha,air}\left[mm\right] = (0.05T_\alpha + 2.85) \bullet {T_\alpha}^{3/2} \quad 4 < T_\alpha \leq 15\ MeV \tag{4}$$

Where T_α is the kinetic energy of the alpha particle in [*MeV*] and $R_{\alpha,air}$ is the range of this alpha particle in the air in [*mm*].

Once the range of the alpha particle in the air $R_{\alpha,air}$ has been calculated, the range of the alpha particle in the VYNS ($R_{\alpha,\ VYNs}$) is given using the Bragg-Kleeman rule to relate the range of the alpha particle in air at normal temperature and pressure to that in the VYNS [9,11,13]:

$$R_{\alpha,VY\acute{N}s} = \frac{\rho_{air}}{\sqrt{A_{ef,air}}} \bullet \frac{\sqrt{A_{ef,VY\acute{N}s}}}{\rho_{VY\acute{N}s}} \bullet R_{\alpha,air} \tag{5}$$

Where ρ_{air} is the density and $A_{ef,\ air}$ is the effective molecular weight of the air at normal temperature and pressure (1.29 kg/m^3 and 14.74 g/mol, respectively). $A_{ef,VYNs}$ is the effective molecular weight of the VYNs, calculated taking into account its composition (mixture of cyclohexanone and poly(vinyl chloride-*co*-vinyl acetate-*co*-vinyl) alcohol)) using the following expression:

$$\sqrt{A_{ef}} = \left(\sum_1^N \frac{\omega_i}{\sqrt{A_i}}\right)^{-1} \tag{6}$$

Where ω_i is the weight fraction of the *i*th element (H, C, Cl and O) that makes up the VYNS and A_i is its corresponding atomic weight.

2.1.3. Range of recoil nuclei in the film

The range of the recoil nuclei in the VYNs ($R_{R,VYNs}$) is determined to establish the minimum film thickness required to stop them and it can be estimated using Eq. (7) [9,11] which, for the same material, relates the range of a recoil particle starting to move at v velocity to that of the alpha particle starting to move at the same v velocity.

$$R_{R,VY\acute{N}s}(v) = \frac{A_R Z_\alpha^2}{A_\alpha Z_R^2} R_{\alpha,VY\acute{N}s}(v) \tag{7}$$

Where A_R and A_α and Z_R and Z_α are the atomic weight and the atomic number of each particle (recoil nucleus and alpha particle, respectively), $R_{\alpha,\ VYNs}\ (v)$ is the empirical function relating the initial velocity v of the alpha particle to its range in the VYNs [cm], and $R_{R,\ VYNs}\ (v)$ is the range in the VYNs [cm] of a recoil nucleus that starts moving at the same initial velocity v. The initial velocity v of alpha and recoil particles is calculated from their kinetic energy.

Equation (5) was used to calculate the range of alpha particles from 1 keV to 1000 keV in the VYNs ($R_{\alpha,\ VYNs}$) and their initial velocity v was calculated from their kinetic energy. The empirical mathematical expression that relates this initial velocity v of the alpha particle to its range in the VYNs ($R_{\alpha,\ VYNs}(v)$) is obtained (equation (8)):

$$R_{\alpha,VY\acute{N}s}(v) = 9 \bullet 10^{-5} e^{2 \bullet 10^{-7} v} \tag{8}$$

When v is in [m/s] and $R_{\alpha,VY\acute{N}s}(v)$ is in [cm].

Using this mathematical expression (Eq. (8)), and following equation (7), the range of recoil nuclei in the VYNS ($(R_{R,\ VYNs}(v))$ is estimated.

It should be noted that equation (7) does not take into account the change in charge state of the recoil particle as it approaches the end of its trajectory [11]. To account for this, the effective charge of the particles (z_{eff}) must be taken into account. The effective charge of the recoil particles analysed ranges between 10 and 8 [12] and that of the alpha particles has values of about 0.2 or even lower for the range of velocities analysed [13], but since in this particular case the effective charge of both particles decreases by the same order of magnitude, there is no difference between taking this effect into account or not.

2.1.4. Alpha particles' energy loss

This calculation has been made because knowing this energy loss makes it possible to correctly identify all the peaks observed in the spectrum, including possible interferences.

Since $E_\alpha = T_\alpha + M_\alpha c^2$, where E_α is the total energy of the alpha particle, T_α is its kinetic energy and $M_\alpha c^2$ is its rest mass. Since the rest mass is constant, $dE_\alpha/dx = dT_\alpha/dx$, and therefore the loss of energy suffered by the alpha particle is that suffered by its kinetic energy.

The total stopping power for alpha particles in the VYNs $(dE/dx)_{VYNs}$, being a mixture of different compounds, is calculated according to equation (9). The total stopping power includes the stopping power due to ionisation and excitation.

$$\frac{1}{\rho_{VY\acute{N}s}}\left(\frac{dE}{dx}\right)_{VY\acute{N}s} = \sum_{i=1}^{n} \omega_i \frac{1}{\rho_i}\left(\frac{dE}{dx}\right)_i \tag{9}$$

Where ω_i is the weight fraction and ρ_i is the density [g/cm^3].of *i*th element (H, C, Cl and O) making up the VYNs. $\left(\frac{dE}{dx}\right)_i$ is the stopping power due to ionisation and excitation for alpha particles in each of the *i* elements making up the VYNs [MeV/m]. This stopping power is determined by equation (10) [9]:

$$\left(\frac{dE}{dx}\right)_i = 4\pi {r_0}^2 z^2 \frac{mc^2}{\beta^2} N_i Z_i \left[ln\left(\frac{2mc^2}{I_i}\beta^2\gamma^2\right) - \beta^2\right] \tag{10}$$

Where r_0 is the classical electron radius [m], mc^2 is the rest mass energy of the electron [MeV]; z is the charge of the alpha particle; N_i is the number of atoms per m^3 of the *i*th element, calculated as $N_i = \rho_i\ N_A/A_i$, where N_A is Avogadro's number and ρ_i, A_i and Z_i are the density [kg/m^3], the atomic weight [kg/mol] and the atomic number [−] of the *i* element, respectively; and I_i is the mean excitation potential [MeV] of it [9]. As for γ and β, they are calculated as follows [9]:

$$\gamma = \frac{T_\alpha + M_\alpha c^2}{M_\alpha c^2}\ [-]\beta = \sqrt{\frac{\gamma^2 - 1}{\gamma^2}}\ [-] \tag{11}$$

Where T_α is the kinetic energy of the alpha particle in [MeV] and $M_\alpha c^2$ is the rest mass of the alpha particle [MeV]. The energy loss suffered by alpha particles as they pass through the VYNs has been calculated for the energy range of interest in this study, from 2 MeV to 10 MeV, the energy of alpha emitters whose activity is usually determined by alpha spectrometry.

2.2. Materials

Recoil contamination generally results from radioactive chains in which short-lived alpha emitters are produced either directly from alpha emitting parents or by decay of short-lived beta emitters; its extent and severity in terms of activity depends on the half-lives of the recoil nuclei and their progeny:

In order to analyse the ability of the alpha source cladding to avoid recoil contamination, 2 matrices with medium and high activity concentrations of Th-228, Th-230 and Th-232 were analysed: ilmenite and soil. These matrices were chosen because Th-228 and Th-229 (used as tracer) are long-lived radioisotopes which, due to the short half-life of their progeny, can cause significant recoil contamination in the detector within a reasonable counting time the. Table 1 shows the activity of each matrix for each isotope.

The activity concentration of the sample is calculated as follows:

$$A = \frac{r_g - r_0}{mR} \tag{12}$$

Where A is the activity concentration of the sample [Bq/kg]; r_g is the gross count rate in the thorium spectral ROI (region of interest) of the sample spectrum [s^{-1}] and r_0 is that of the same ROI's of the background spectrum [s^{-1}]; m is mass of the test-portion mass [kg] and R is the total yield of the radiochemistry and measurement process [−], which is calculated as follows:

$$R = \frac{r_{At,g} - r_{At,0}}{A_t} \tag{13}$$

Where $r_{At,g}$ is the gross count rate observed in Th-229 spectral ROI of the sample spectrum [s^{-1}] and $r_{At,0}$ is that of the same ROI's of the background spectrum [s^{-1}] and A_t is the activity of the tracer added to the sample [s^{-1}] when the radiochemical separation process is carried out.

The combined standard uncertainty has been calculated following the guide to the expression of the uncertainty in measurement of ISO/IEC Guide 98 [14]:

Fig. 1. Connecting 13 batteries in series to obtain 39 V and later connecting them in parallel to the base.

$$u(A) = \sqrt{\left(\frac{r_g}{t} + \frac{r_0}{t_0}\right)\cdot\left(\frac{1}{m \bullet R}\right)^2 + A^2 \bullet \left[\left(\frac{r_{At,g}}{t} + \frac{r_{At,0}}{t_0}\right) \Big/ \left(r_{At,g} - r_{At,0}\right)^2 + u_{rel}^2(A_t) + u_{rel}^2(m)\right.} \tag{14}$$

Where u(A) is the uncertainty of the activity concentration [Bq/kg]; t and t_0 are the sample and background counting times [s]; $u_{rel}(A_t)$ and $u_{rel}(m)$ are the relative uncertainties of the activity of the tracer added to the sample and that of the mass, respectively.

For each matrix, 3 aliquots were prepared and measured, all of them in the same detector, a Canberra alpha particle spectrometer equipped with 450 mm^2 passivated implanted planar silicon (PIPS) detectors, whose energy and efficiency calibration is established using a standard electroplated calibration source of 96.5 Bq; the efficiency obtained is 28.52 % and its background is of $5.8{\cdot}10^{-5}$ cps. Measurements were performed for 300000 s after establishing a vacuum of 0.1 mm Hg in the chamber and at a detector-source distance of 5 mm.

Six spectra were recorded for each sample: in the first, a thin VYNS film covers the sample to avoid recoil contamination; then, after removing the film, four potential differences of 3, 6, 18 and 39 V are applied to the plate on which the alpha source is deposited; finally, to quantify the effect of both methods (covering the alpha source (CS) and applying a negative potential (NP)), a measurement is made on the sample without applying any system to avoid recoil contamination (Source). The detector on which the experimental measurements were carried out can provide a potential difference of 3V using 4 batteries connected in parallel to apply this negative potential uniformly to the base. To apply the other voltages (6V, 18 V and 39 V), the number of batteries required to obtain the desired potential difference were connected in series and then in parallel with the base, as is shown in Fig. 1 (connection of batteries to apply 39 V).

Table 1
Activity concentration [Bq/kg] of Th-228, Th-230 and Th-232 for each matrix. This table also shows the uncertainty of this activity (k = 2).

MATRIX	Activity ± Uncertainty [Bq/kg]		
	Th-228	Th-230	Th-232
ILMENITE	177,9 ± 13,7	108,6 ± 8,9	343,7 ± 25,2
SOIL	24,7 ± 2,5	46,5 ± 3,9	44,2 ± 3,6

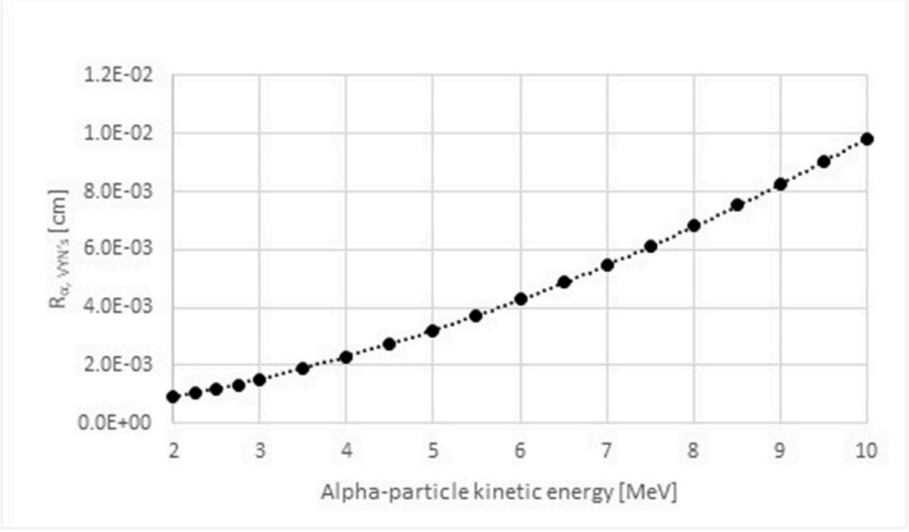

Fig. 2. $R_{\alpha,\ VYNs}$ alpha particle mean range into the VYNs [cm] as a function of its kinetic energy [MeV].

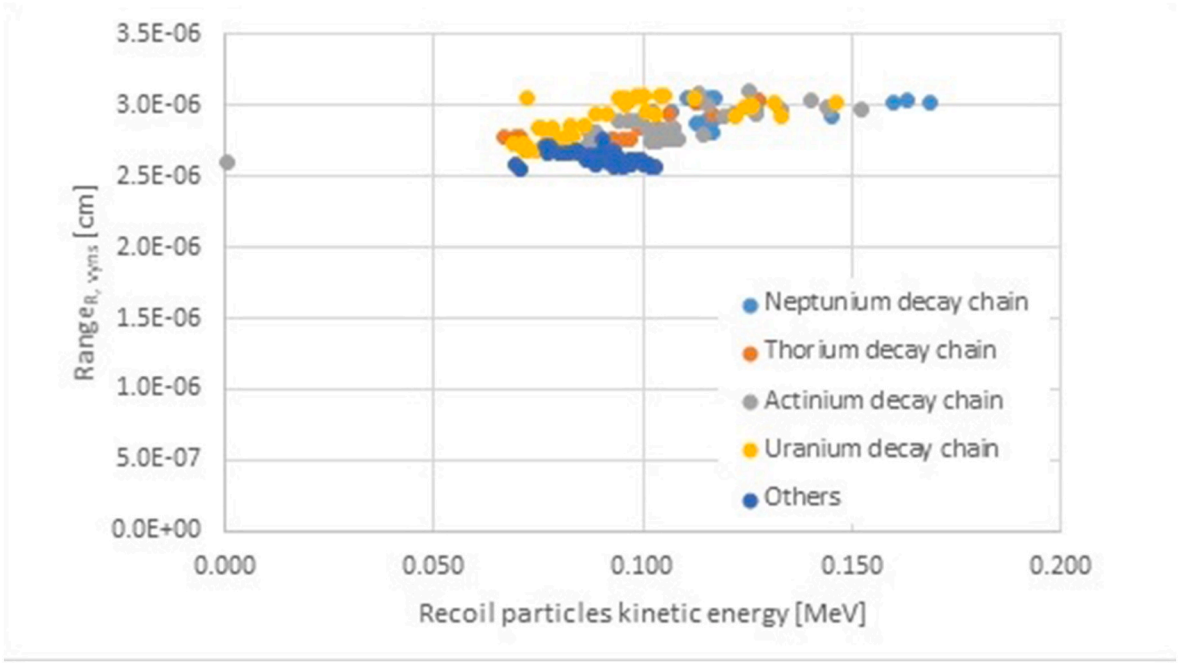

Fig. 3. The range of the recoil nuclei range in the VYNs ($R_{R,\ VYNs}$ [cm]) as a function of the kinetic energy [MeV] of the recoil particles.

3. Results and discussion

3.1. Range of alpha particles in the film

The range values of the alpha particles in the film range from $9.03 \cdot 10^{-4}$ cm for alpha particles of 2 MeV–$9.81\ 10^{-3}$ cm for 10 MeV.

Fig. 2 shows the range of alpha particles in the VYNs ($R_{\alpha,\ VYNs}$ [cm]) as a function of their kinetic energy.

3.2. Range of recoil nuclei in the film

The kinetic energy values of the recoil nuclei of the analysed alpha-emitting radioisotopes, i.e. those emitting by alpha particles between 2 and 10 MeV, range from 0.001 to 0.168 MeV. The value of the range of the recoil nuclei in the VYNs is calculated using equations (7) and (8), and the maximum value of the thickness required to stop the recoil nuclei is slightly more than $3 \cdot 10^{-6}$ cm of film, as we can see in Fig. 3, but lower than $3.5 \cdot 10^{-6}$ cm.

This Fig. 3 shows the range obtained for the alpha emitters of the neptunium, thorium, actinium and uranium decay chains and for the artificial alpha emitters (Others).

3.3. Amount of VYNs required to prevent recoil contamination but to allow the passage of all alpha particles

The film required to cover the source must be uniform and as fine as possible. However, from a practical and routine point of view, it is quite complicated to produce these films over the source thinner than 60 μg/cm^2, with typical values between 60 and 70 μg/cm^2. Therefore, these mass thicknesses, corresponding to a thickness in the range ($4.41 \cdot 10^{-5}$ – $5.90 \cdot 10^{-5}$) cm, are considered a realistic value for this parameter.

This thickness is smaller than the minimum range of alpha particles

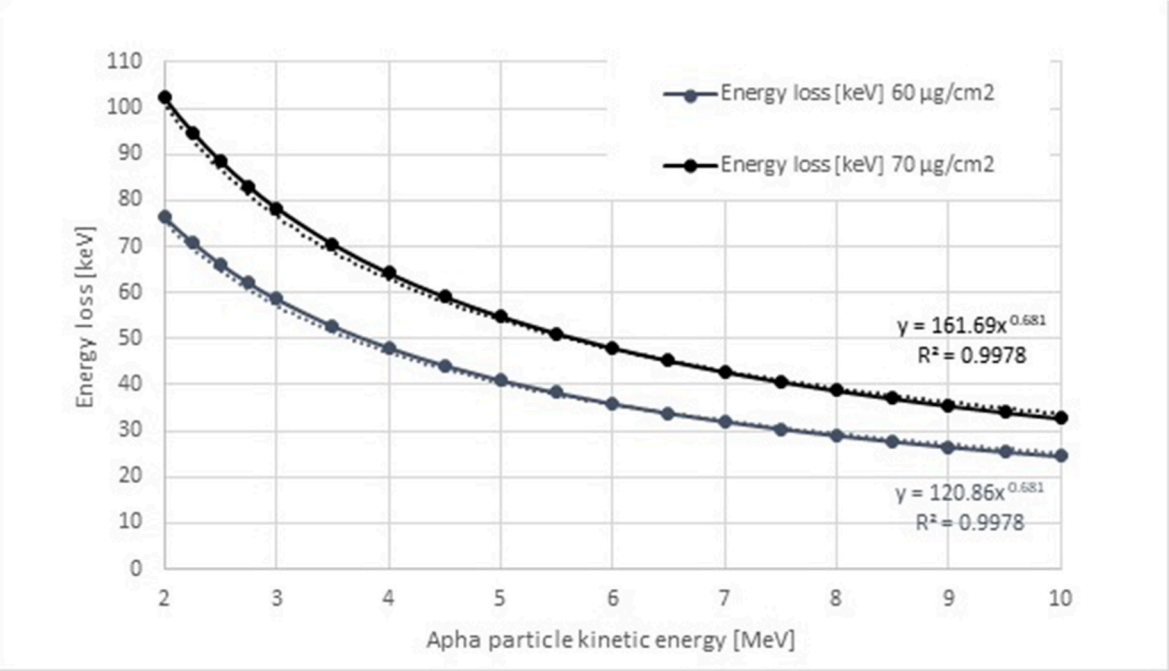

Fig. 4. The energy loss of the alpha particle [keV] as it passes through the film, for 60 and 70 µg/cm^2, as a function of its kinetic energy [MeV].

in the VYNs (9.03·10-4 cm), so it can be concluded that all emitted alpha particles will reach the detector and therefore the detection efficiency is not affected by the use of the film. Furthermore, since the maximum range of the recoil nuclei in the VYNs ($3.5{\cdot}10^{-6}$ cm) is an order of magnitude smaller than the film thickness, the recoil particles will not pass through the film. It is concluded that a thickness between 4.41·10-5 cm and 5.90·10-5 cm meets both objectives.

3.4. Energy loss of alpha particles crossing the film

Fig. 4 shows the energy loss of alpha particles as a function of their kinetic energy, when passing through the VYNs film for the two typical values considered for the thickness of this film, 60 and 70 µg/cm^2. As can be seen in the figure, the expression relating the loss of the energy [keV] to the kinetic energy of the alpha particle [MeV] is a power function, not linear and is useful for identifying any interference that may appear in the spectrum. The fitting of these equations is also shown in the figure.

The energy loss suffered by the alpha particles as they pass through the film will shift all the peaks obtained in the spectrum towards lower energies, but this shift will not be the same for all the peaks, as it depends on the energy of the alpha particles and the mass thickness of the VYNs. For example, the value of the shift observed for 4 MeV varies between 48 and 64 keV (and for 8 MeV between 28 and 38 keV) for the two mass thicknesses considered.

3.5. Activity measurements

First, 6 alpha sources covered with the film VYNs are measured for 300000 s. The background is measured for 600000 s.

Table 2 shows the activity concentration and its uncertainty (coverage factor $k = 2$) [Bq/kg] obtained when measuring ilmenite and soil samples after covering the source with a foil (CS), after applying a negative potential after removing the foil (NP) and finally after measuring the source without covering it or applying any negative potential (SOURCE). These activity concentrations and their uncertainties have been calculated using equation (12 13 and 14). In this first case, the negative potential applied to the base on which the source is placed (3 V) is that specified by the commercial equipment used to carry out the experimental measurements. This negative potential is obtained by connecting four batteries in parallel to the base.

The results in Table 2 show that the two methods used to avoid recoil contamination may be more or less effective, but they do not affect the evaluation of the activity present in the sample, since, taking into account their uncertainties, there is an overlap in the value of the activity concentration.

To see the effect of both methods on the peak shape, the FWHM (Full Width at Half Maximum) and FWTM (Full Width Tenth Maximum) of

Table 2
Activity concentration and its uncertainty [Bq/kg] of Th-232, Th-230 and Th-228 for ilmenite and soil.

	ILMENITE			SOIL		
	SOURCE	CS	NP	SOURCE	CS	NP
Th232	327.1–23.83	314.7–22.58	323.7–23.72	42.53–3.745	41.78–3.785	42.79–3.909
Th230	106.2–8.604	96.96–7.832	98.24–8.141	41.48–3.745	44.91–4.00	42.62–3.898
Th228	171.5–13.15	166.8–12.58	162.9–12.64	23.85–2.508	24.86–2.597	22.18–2.440
Th232	320–20.83	354.9–23.23	325.1–44.32	41.92–3.437	45.92–3.798	43.53–3.648
Th230	98.06–6.985	115.4–8.198	110.9–14.07	42.67–3.487	48.44–3.963	43.00–3.614
Th228	163.0–11.04	180.2–12.27	163.4–22.51	21.67–2.105	24.33–2.360	22.75–2.256
Th232	322–26.25	349.2–28.50	339.5–29.20	40.38–3.556	44.60–3.683	43.10–3.633
Th230	108.3–9.997	104.3–9.779	104.8–10.40	42.66–3.406	45.78–3.761	42.67–3.607
Th228	165.3–14.36	182.0–15.75	165.2–15.28	22.67–2.226	24.80–2.373	21.84–2.205

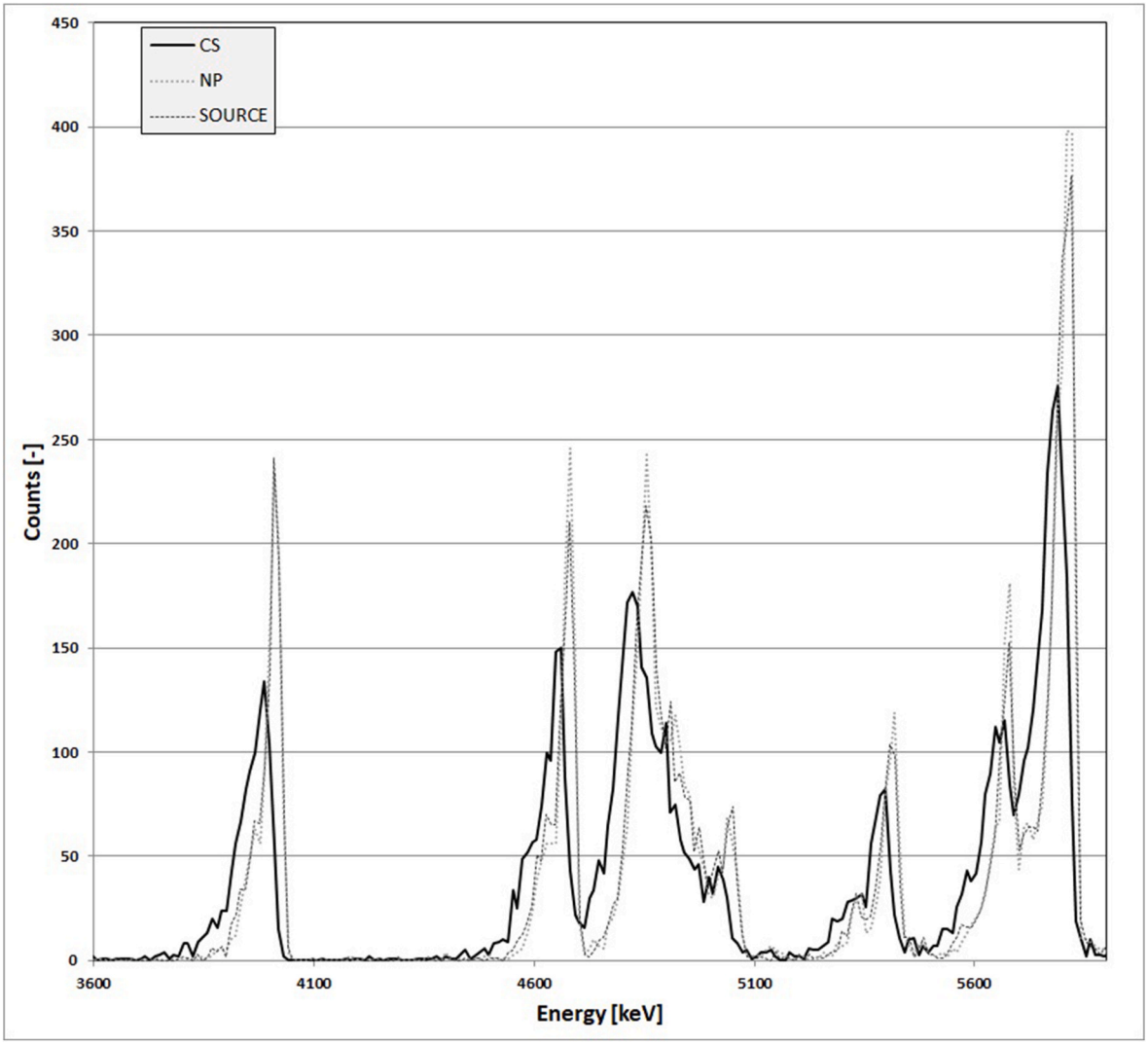

Fig. 5. The spectra obtained for SM131TH for three realised measurements: SOURCE, CS and NP.

each peak are measured directly from the spectra The result shows that, covering the source significantly increases the FWHM values, on average of 61 % higher, for example in the case of soil where the peaks of Th-232, Th-230, Th-229 and Th-228 change from 35 keV to 66 keV, from 37 keV to 56 keV, from 77 keV to 108 keV, and from 35 keV to 53 keV respectively, whereas using a negative potential has no effect. As far as the FWTH is concerned, it is observed that this value increases by an average of 28 % when the source is covered, whereas there is hardly any change in its value when a potential difference is applied to the source. This expected result is due to the energy loss of the alpha particles as they pass through the film. However, even in this case, the peaks observed are sufficiently well defined to be analysed (Fig. 5).

Fig. 5 shows the spectra obtained for the same source of soil (SM131TH): without covering the source or applying a negative potential (SOURCE), after covering the source with a film (CS) and after applying 3 V (NP).

Only the region where the isotopic peaks of Th-230, Th-229, Th-228 and Ac-225, a descendant of Th-229, appear in the spectra, to see that they are indeed perfectly distinguishable from each other.

With regard to the number of counts collected under each peak, a similar number of counts has been collected in each isotopic peak of the three measurements; therefore, the activity evaluated from these peaks in the three measurements is equivalent (Table 2). With regard to the difference observed in the number of counts representing the maximum point of each peak, it is observed that the lowest number is recorded in the measurement of the source made after the covering (CS); this is due to the loss of energy that the presence of the film causes in the alpha particles emitted by the source, causing the peaks to broaden in spite of

Table 3
Counts [−], FWHM [keV] and FWTM [keV] of Th-232, Th-230, Th-229 and Th-228 for SMTH131.

	SOURCE	COUNTS [−]		NP	SOURCE	FWHM [keV]		NP	SOURCE	FWTM [keV]		NP
		CS				CS				CS		
Th-232	1056	1022		1003	32	73		39	114	163		108
Th-230	1034	1100		1001	38	57		36	115	171		117
Th-229	2493	2435		2352	74	124		60	295	331		271
Th-228	592	611		523	42	50		46	157	171		151

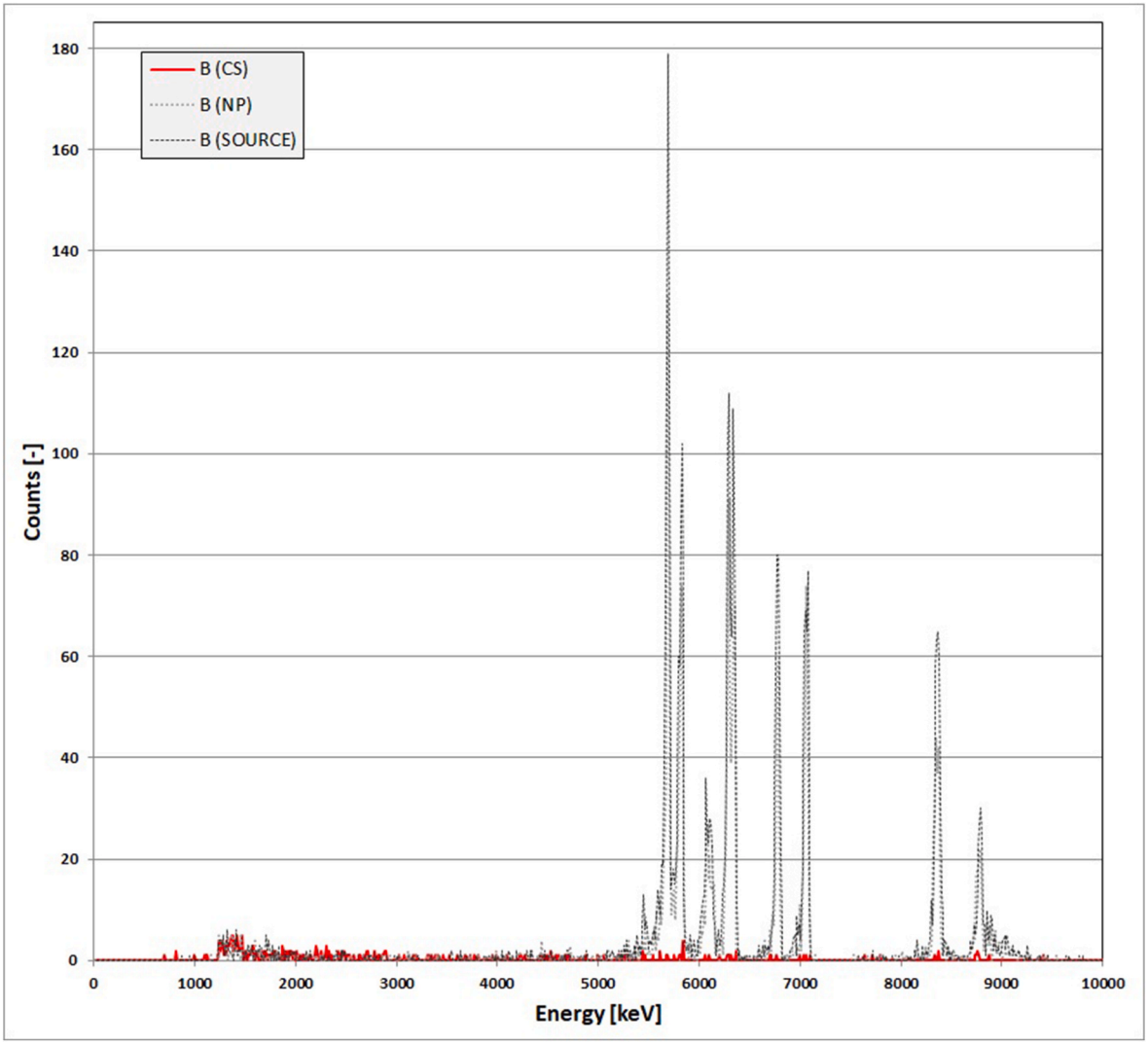

Fig. 6. Background after measuring all the sources covered with a film (B (C)S), after measuring the sources without using a film but applying 3V negative potential (B (NP) and after measuring the sources without using a film or applying a negative potential ((B (source)).

having a similar number of counts. Table 3 shows the number of counts [−], FWHM [keV] and FWTM [keV] obtained when measuring SM131TH in the three different ways defined.

Fig. 6 shows the spectra obtained for the backgrounds recorded during 600000 s after carrying out the measurements corresponding to each group of samples. The first one shows no recoil contamination after measuring the sources covered with the film (B (CS)), the second one shows the background obtained after measuring the samples with a relative potential of 3 V applied to the source (B (NP)), and the last one after measuring the source without covering it or applying a negative potential (B (SOURCE)).

These three backgrounds were measured in the same detector and under the same vacuum conditions (0.1 mm Hg) for a counting time of 600000 s; therefore, after each series of sample measurements, the detector was left at rest for the time necessary for the contamination to disappear (until the number of counts recorded in the spectra was equal to the usual number of counts in the spectra of the backgrounds). The detector has thus been dedicated to this work for just over two years.

As it can be seen in Fig. 6, the spectrums of the backgrounds obtained were different. If we analyse the number of counts recorded in the same ROI in the 3 spectrums (from channel 353 (3836.5 keV) to 861 (9343.7 keV)), the region in which the counts resulting from the recoil nuclei appear, we get the following.

- In the spectrum obtained after covering the source, 69 counts were recorded (Fig. 6 (B(CS)); in that obtained after applying a negative potential of 3 V, 2873 counts were recorded (Fig. 6 (B(NP)); and, in the last one, that obtained after measuring the sources without covering them or applying a negative potential, 4074 counts were recorded (Fig. 6 (B(SOURCE)).

It can be concluded that covering the source avoids recoil contamination. Regarding the effect of applying a 3 V of negative potential to avoid recoil contamination, even if its application reduces this type of contamination, it has not eliminated it. In order to analyse how increasing the voltage could improve the performance of this system, the voltage required to prevent the recoil nuclei from reaching the detector was analysed theoretically, and then experimental tests were carried out at increasing voltages of 6, 18 and 39 V.

Theoretically, an approximation of the potential required (ΔV) to stop 100 % of the recoil nuclei can be calculated from Eq. (15),

$$\Delta V(V) = \frac{T_R}{Z_R} \tag{15}$$

Where ΔV is the potential required to stop the recoil nucleus, T_R is the kinetic energy of the recoil nucleus [eV], which values between 0.07 MeV and 0,17 MeV; and z_R [−] is its atomic number. Applying this

formula to the particular case of thorium samples, the values obtained range from 800 to 3000 V, much higher than those used in the experiments did.

The calculated values are the lowest because it is assumed that the charge of the recoil nucleus does not change. If we took this change into account and used Z_{eff} instead of Z_R, this value would be much higher [12]. This result is in agreement with Sill and Olsen [1], who stated by that at short distances and pressures below 0.1 mm Hg, the application of even several hundred volts does not produce a significant reduction in contamination in recoil contamination.

From a practical point of view, it is not realistic to think of applying this voltage to the base supporting the alpha source, so we experimentally analysed how much contamination by recoil nuclei could be reduced by applying increasing but routinely achievable voltages during routine use of the equipment: 6, 18 and 39 V. As in the case of the 3 V, these measurements were carried out under low vacuum conditions (0.1 mm Hg), with the source positioned 5 mm from the detector.

The maximum potential value is 39 V because, at most, in our detector and for its use in routine measurements, a maximum negative potential of 39 V could be applied using 13 batteries in series connected in parallel on the base (see Fig. 1).

In order to observe the evolution of the number of counts recorded in the background spectra as a function of the applied voltage, Fig. 7 shows the number of counts recorded for each of the voltages applied, starting with the value obtained without applying any voltage (4074), bearing in mind that a minimum of 800 V would be required to retain these recoil nuclei. The trend observed in this figure fits well with an exponential. This trend is consistent with the experimental results observed by Sill and Olsen [1]: the application of even several hundreds of volts did not lead to a significant reduction in this type of contamination.

As can be seen from this figure, and as it was expected, the higher the voltage, the greater the reduction in recoil nuclei contamination. However, even in the best of the case studied, 39V, the reduction is only up to 44 % compared to doing nothing to avoid it.

If we compare the spectra obtained after measuring the source in the three defined ways, we can see that.

- When the source is covered with the film (CS), the number of counts recorded by the detector is in some cases slightly higher than that presented by the source alone (SOURCE), and in other cases slightly lower; on average, the latter is obtained to be % 1 higher. This difference could be explained by the statistical counting error inherent in counting. This fact confirms that, as theoretically demonstrated, the use of the film does not prevent all the alphas emitted by the source from reaching the detector. When a negative potential (NP) of 3 V is applied, the number of counts recorded by the detector is on average 8 % lower, which cannot be due solely to the counting error. As the value of the applied negative potential increases, the number of counts recorded decreases, so that when a voltage difference of 39 V is applied, the average number of counts recorded is actually 16.5 % lower than when measuring the source alone (SOURCE).
- Regarding the resolution of the peaks observed in the spectra (FWHM), when the source is covered with the VYNs (CS), this parameter is always higher, showing an average of 61 % higher with respect to that observed when no system is applied to avoid recoil contamination (SOURCE); nevertheless, as can be seen in Figure (5), there is no problem in distinguishing the peaks between them. On the contrary, when different voltages are applied to the source (NP), the values obtained in terms of the resolution show a very low variability, the values remain almost the same; in fact, the resolution is on average 1 % lower when 3 V is applied g, and 0.5 % lower when 39 V are applied,.

Other equipment commonly used to perform alpha measurements, Alpha Analyst, have an automatic recoil suppression system: a user-defined air thickness (between 12 and 16 g/cm^2) coupled to a 5 V potential. After activating this recoil suppression system, the vacuum in the housing decreased, but after a few hours of counting, the vacuum increased to 7 mm Hg. The range of recoil particles inside was also estimated to be between 3.25 mm and 4 mm, close to 5 mm, the distance used in the laboratory to perform an activity measurements. It should be remembered that the calculated range is the mean range and not the extrapolated one, since the difference between them is about 5 %; therefore, this system cannot be guaranteed to prevent contamination of the detector by recoil nuclei for such close distances to the detector. It should also be noted that at these vacuum pressure values, the efficiency of the detector is strongly reduced, since the number of alpha particles that would reach the detector would also be greatly reduced; in fact, these devices usually have as reference values pressures lower than 0.5 mm Hg. We can therefore conclude that this contamination reduction system could only work for very short counting times, so that the vacuum would remain below atmospheric pressure for approximately less than a day.

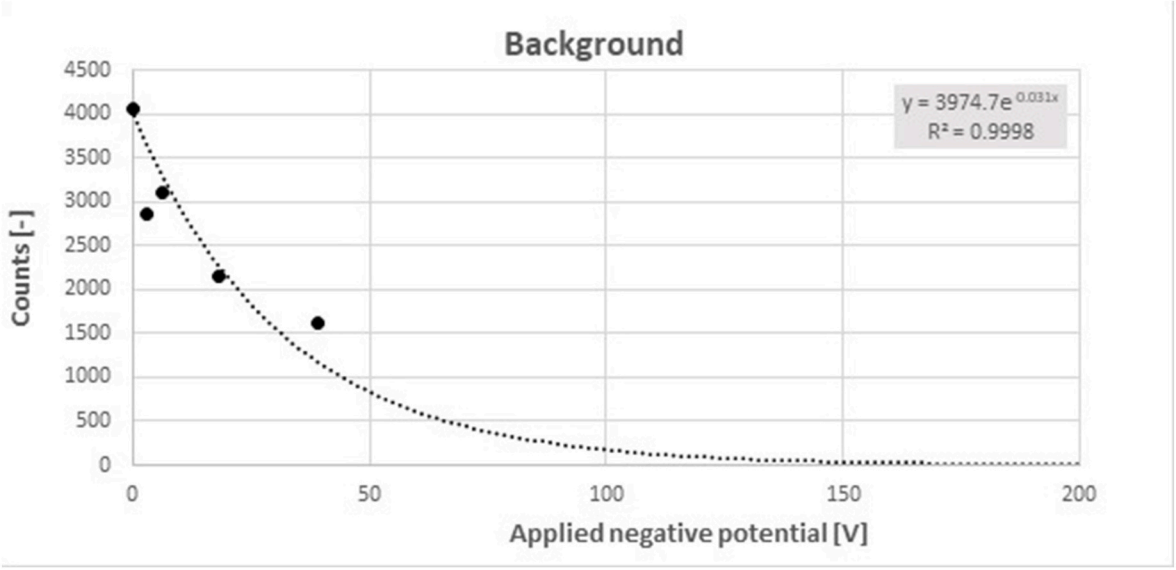

Fig. 7. Background obtained after measuring all samples without using film but applying 3V, 6 V, 18V and 39 V.

4. Conclusions

Experimental measurements, carried out under low vacuum conditions (0.1 mm Hg) and with the source placed very close to the detector (at 5 mm), confirm the theoretical calculation regarding the effectiveness of the VYNs film in avoiding recoil contamination and the effect of its use on the recorded activity:

Based on the analytical calculation carried out to obtain the range of the recoil particles in the VYNs when performing alpha spectrometry, it is found that for the most common alpha emitters that this range, less than $3.5{\cdot}10^{-6}$ cm, is smaller than the thickness of the film which is used to cover the source (between $4.41{\cdot}10^{-5}$ and $5.90{\cdot}10^{-5}$ cm). The experimental background obtained after measuring the sources with this film confirms that this film is very effective in avoiding detector contamination. Again, based on the analytical calculations carried out, the use of the film (VYNs) does not reduce the number of the alpha particles that can be recorded by the detector since the small range of alpha particles in the film, $9.03{\cdot}10^{-4}$ cm, is greater than its thickness; the number of counts obtained from the spectra after measuring the sources without and with the film confirms this fact, since the difference observed in the number of counts recorded for each peak can be explained by the counting error. In addition, the mathematical expression relating the energy loss of the alpha particle when passing through the film to its energy allows the identification of any interference that may be observed in the spectrum. Finally, the effect that the presence of the VYNs could have on the resolution was also studied and, although the spectra obtained show that it is much higher than that corresponding to the source without covering, the peaks observed remain very distinguishable.

This work also tested the most common systems for suppressing recoil contamination used by different measuring commercial equipment: applying 3 V to the source and creating an air cushion together with 5 V. Experimental measurements, also carried out for laboratory measurement conditions, show that applying 3 V to the source does not prevent recoil contamination. The effect of increasing the negative potential applied to the source on the ability to avoid recoil contamination has also been studied using different potential values; the results obtained show that the value required to almost completely eliminate recoil contamination is too high, around several hundred of volts, which is very difficult to establish and maintain over a long period of time for routine measurements. As for the second method, it does not meet the needs of our laboratory in order to obtain the low LD values sometimes required, which are obtained by measuring samples under low vacuum conditions for several days.

Since the experimental measurements were carried out in low vacuum conditions (0.1 mm Hg) and with the source very close to the detector (at 5 mm), this covering proved capable of protecting the detector against contamination by nuclei, even in these conditions, without the need to combine the use of this covering with other additional measures commonly used, such as the application of a potential difference, combined or not with the creation of an air cushion obtained by applying a certain pressure in the detector chamber, which facilitates the handling of the detector during the measurements. It was also found that the integrity of the VYNs film used to cover the source was maintained over time.

It can therefore be concluded that the type of film used to prevent recoil contamination achieves this objective without blocking the arrival of all the alpha particles emitted by the source and the detector. It is therefore confirmed that the use of these VYNs makes it possible to keep the detector free from recoil contamination, especially when measuring samples with high activity, where the probability of contamination of the detector by recoil nuclei is higher, and even when the sample remains inside the detector for a long time without any possibility of removal, as was the case during the pandemic. This security of measurement is of vital importance, since it not only prolongs the use of the detector over time, but also guarantees that the measurements made are correct, since there are no interferences that affect the correct determination of the peaks of interest; likewise, keeping the detector background as low as possible makes it possible to achieve the low LD values established by the competent authorities, both in the surveillance plans carried out around the nuclear power plants that are in operation and in the subsequent decommissioning process that may be carried out.

During the preparation of this work the author(s) used DeepL Translate in order to to check English writing. After using this tool/service, the author(s) reviewed and edited the content as needed and take(s) full responsibility for the content of the publication.

CRediT authorship contribution statement

C. Olondo: Conceptualization, Formal analysis, Investigation, Methodology, Supervision, Validation, Writing – original draft, Writing – review & editing, Visualization. **R. Idoeta:** Formal analysis, Methodology, Supervision, Validation, Visualization, Writing – review & editing. **M. Herranz:** Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Supervision, Validation, Visualization, Writing – original draft, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

References

[1] C.W. Sill, D.G. Olson, Sources and prevention of recoil contamination of solid-state alpha detectors, Anal. Chem. 42 (13) (1970) 1596–1607.
[2] K.G.W. Inn, E. Hall, J.T. Woodward, B. Stewart, R. Pollanen, L. Selvig, S. Turner, I. Outola, S. Nour, H. Kurosaki, J. LaRosa, M. Schultz, Z. Lin, Z. Yu, C. McMahon, Use of thin collodion films to prevent recoil-ion contamination of alpha-spectrometry detectors, J. Radioanal. Nucl. Chem. 276 (2) (2008) 385–390.
[3] K.G.W. Inn, Thin film to prevent recoil contamination of solid-state alpha detectors, in: Proceedings of the Sixth International Conference on Methods and Applications of Radioanalytical Chemistry, Kailua-Kona, Hawaii, 2003. April 7-11.
[4] J.L. Gels, J.T. Elliston, J.W. Talnagi, S.E. Glover, H.B. Spitz, A pre-concentration neutron activation analysis method for the determination of 232Th utilizing a dry-tube irradiation facility, J. Radioanal. Nucl. Chem. 277 (1) (2007) 217–221.
[5] N. Vainblat, O. Pelled, U. German, G. Haquin, A. Tshuva, Z.B. Alfassi, Determination of parameters relevant to alpha spectrometry. De termination of parameters relevant to alpha spectrometry when employing source coating, Appl. Radiat. Isot. 61 (2004) 307–311.
[6] J. Janda, P. Sládek, D. Sas, Electrodeposition of selected alpha-emitting radionuclides from oxalate-ammonium sulfate electrolyte and measured by means of solid-state alpha spectrometry, J. Radioanal. Nucl. Chem. 286 (2010) 687–691.
[7] C. Cosma, O.A. Rusu, V. Cosma, D. Nita, R. Cs Begy, A. Timar Gabor, A. Astilean, Protection of alpha spectrometry detectors using thin formvar films and influence on detection characteristics, IEEE Trans. Nucl. Sci. 59 (2012) 1175–1179.
[8] A. Vander der Wijk, L. Venema, S.P. Steendam, The use of thin film plastic foils in low-level alpha spectrometry, International Journal of Radiation Applications and Instrumentation, Part A, Appl. Radiat. Isot. 38 (1987) 1061–1065.
[9] N. Tsoulfadinis, S. Landsberger, Measurement and Detection of Radiation, third ed., Taylor and Francis Group, New York, 2011.
[10] Nuclide Lara, 2022. http://www.nucleide.org/Laraweb/index.php.
[11] G.F. Knoll, Radiation Detection and Measurement, second ed., John Wiley and Sons, Inc., New York, 1989.
[12] A. Stoklosa, J. Zajecki, S.S. Kurek, Effective nuclear charge of an ion, Materials Science-Poland 22 (I) (2004) 35–45.
[13] R.D. Evans, The Atomic Nucleus, first ed., McGraw-Hill Inc., New York, 1955.
[14] GUM, Evaluation of measurement data – guide to the expression of uncertainty in measurement, JCGM 100 (2008) 2008.