Incrustation of α -particle emitters in the source backing: influence on activity measurements

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Abstract. Alpha particles emitted from radioactive sources are often measured using a 2π counting geometry in order to determine the activity with a low deviation. The ratio $C_{2\pi}/A$ (counting rate/activity) can, however, deviate from the theoretical value of 0.5 because of backscattering in the backing material, scattering and absorption of alpha particles in the source. The experimental counting rates are, therefore, corrected for these effects (backscattering for all sources, plus self-absorption for sources of non-negligible thickness) to determine the real source activity. However, the corrections needed for situations corresponding to alpha-particle sources in which the radionuclides are not deposited but incrusted in the backing material have not been considered. The aim of the present work was therefore to study the influence that incrustation in the backing can have on the total detection efficiency, and hence on the activity estimated for the source. To this end, we used the Monte Carlo computer code SRIM to model the behaviour of the alpha particles in the backing material.

Key words: alpha-particle spectrometry • Monte Carlo (MC) simulation • SRIM

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Introduction

Alpha particles sources are commonly standardized by using ionization chambers in a 2π counting geometry, because the theoretical efficiency is known and the activity can be determined with low deviation. However, the efficiency $C_{2\pi}/A$ (counting rate/activity) can deviate from the theoretical value of 0.5 because some of the alpha-particles initially emitted downwards towards the backing material may be backscattered into the 2π detector, while other particles emitted upwards towards the detector are scattered and/or absorbed in the source. For this reason, some corrections must be made to the experimental counting rates in order to determine the real source activity. These corrections have been well studied for sources of negligible thickness [2, 4, 8, 12] where backscattering is the only correction needed, and also for sources with a non-negligible thickness of the radioactive deposit on the backing [5–7, 9–11, 13], in which additional corrections are required because of the self-absorption of alpha particles in the source substrate. All the above studies were performed assuming Gaussian scattering models or using Monte Carlo (MC) procedures to simulate the interaction of alpha particles in the source.

However, the corrections needed for situations corresponding to alpha-particle sources where the radionuclides are not deposited but incrusted in the backing material have not been considered. This is the case, for example, of the alpha-particle sources prepared by electrodeposition, in which the fine deposits minimize the effects of self-absorption, but in which the radionuclides may penetrate into the source support. For this type of source, the counting rate $C_{2\pi}$ will be less than that for sources with radionuclides deposited superficially onto the backing.

The aim of this work was to study the influence that the incrustation of nuclides in the backing may have on the total detection efficiency, and hence on the activity estimated for the source. To model the behaviour of the alpha particles in the support material, we used MC simulation, in particular, the well-established Monte Carlo computer code SRIM [14]. This was developed to simulate the transport of ions in matter, and here we applied it to study the corrections needed in those cases where alpha-particle emitters are incrusted in the source's backing.

The values of the total detection efficiencies $C_{2\pi}/A$ were obtained for sources with three backing materials with very different atomic numbers: Al, Ag, and Pt. For each backing, several alpha-particle energies were considered, together with a wide range of incrustation depths in the source support.

Monte Carlo simulation

We used the latest version available of the Monte Carlo code SRIM [15]. The simulation follows the path of charged particles through all types of materials, and computes their energy losses as well as the scattering along their paths. The program takes an input file containing the initial characteristics (position, direction cosines, and energy) of the particles in the source. Once all the events have been simulated, the program generates files containing the energy, position, and direction of the ions emerging from the front or from the back of the source. It can also provide data on recoil, sputtering, radiation damage, etc.

For this study, we considered three backings with very different atomic numbers (Al, Ag, and Pt) which are commonly used in the deposition procedures for alpha emitters, and three values for the energy of the alpha-particles (3, 5, and 7 MeV). For each alpha-particle energy and backing, we simulated three different cases (Fig. 1):

- a) Source with a radioactive substrate of negligible thickness located just on the surface of the backing (surface deposition).
- b) Source with alpha particles emerging from a given depth x in the backing (nuclides incrusted at a given depth x).
- c) Source with the radionuclides uniformly distributed in the backing, from the surface down to a depth *x* (nuclides incrusted uniformly down to a depth *x*).

For each individual case, we carried out two simulations, each including 20 000 alpha particles and different random number seeds. As required for SRIM, input files were prepared containing the incident direction cosines and spatial coordinates for each alpha particle. The isotropic sources were then simulated and the tracks of the alpha particles in each backing material were followed.



Fig. 1. Scheme of the three cases of alpha-particle sources considered: (a) deposition on the backing surface, (b) nuclides incrusted at a single given depth x within the backing, and (c) nuclides uniformly incrusted from the surface down to a depth x in the backing.

Results and discussion

Sources with a negligible substrate thickness deposited on the backing

First, we simulated alpha-particle sources in which the radioactive substrate has a negligible thickness and is located just on the surface of the backing (surface deposition). In this case, there is no self-absorption, so that the correction to the counting rate is only due to backscattering of alpha-particles in the backing.

Figure 2 shows the energy distribution obtained for the backscattered alpha particles from Al, Ag, and Pt backings, in the particular case of 5 MeV. The maximum of this distribution corresponds to particles emerging from the backing with energies close to zero. It corresponds to particles which have undergone many weak collisions in the material (multiple scattering). A second dominant structure is found for particles with energies somewhat less than the initial energy, which can be attributed to particles undergoing a few weak collisions (plural scattering). This behaviour is coherent with Crawford's theoretical treatment [2] in which the backscattering is mainly due to small-angle scattered particles, i.e., primarily multiple scattering as a result of a large number of weak electronic collisions.

The $C_{2\pi}/A$ ratios were evaluated dividing the number of particles leaving the source upwards towards the 2π



Fig. 2. Energy distributions of backscattered particles for sources with the nuclides deposited superficially onto backings of Al, Ag, and Pt (zero thickness). The distributions correspond to alpha particles of 5 MeV and are represented with 10 keV energy intervals.

sr detection chamber by the number of particles emitted isotropically in all directions (20 000 in this work). Table 1 gives the values obtained by simulation for these ratios in the sources with a radioactive substrate



Fig. 3. Spectra of detected alpha particles for the particular case of 5 MeV, when the radionuclides are incrusted as in situation type (b) at a depth of $50 \ \mu\text{g/cm}^2$ in a backing of Pt (233 Å), and in the case when they are incrusted as in situation type (c) uniformly down to this same depth. The spectra are represented with 10 keV energy intervals.

of negligible thickness located just on the surface of the backing. Each value corresponds to the mean of two simulations, each including different random number seeds. The corresponding uncertainties were obtained by assuming the standard deviation as an estimation of the dispersion. As expected, the $C_{2\pi}/A$ values for each backing decrease gradually with alpha-particle energy. In addition, the $C_{2\pi}/A$ ratio (and consequently the corresponding backscattering coefficient) increases with atomic number Z of the backing, as has already been noted by other workers [3, 4].

Sources with nuclides incrusted at a depth x

Second, we simulated sources where the alpha-particle emitters are embedded in the source backing at a given depth x. Figure 3 shows the spectrum of detected alpha particles for the particular case of 5 MeV, and an incrustation depth of 50 µg/cm² in a backing of Pt (233 Å). As in the previous case, the $C_{2\pi}/A$ ratios were evaluated dividing the total number of alpha particles detected in 2π sr by the number of initial particles emitted isotropically from the source.

Figure 4 shows the ratios C(x) between the $(C_{2\pi}/A)_x$ values for these sources (nuclides incrusted at a depth x) and the values of $(C_{2\pi}/A)_0$ for sources with the nuclides deposited on the surface. One observes that, for a given backing and alpha-particle energy, the C(x) values deviate more from unity as the incrustation depth increases. This means that the deviation in the activity determination if the incrustation is not considered would increase with the depth of the nuclide incrustation. This is due to the absorption in the backing of particles initially emitted "towards" the detector and also to the backscattered alpha particles.

Table 1. Calculated values of the ratio $(C_{2\pi}/A)_0$ for alpha-particle sources with a substrate of negligible thickness deposited superficially on the backing material. The uncertainties correspond to one standard deviation

Energy (MeV)	Energy (MeV) Al		Pt	
3	0.5093 ± 0.0002	0.5230 ± 0.0005	0.5348 ± 0.0002	
5	0.5074 ± 0.0002	0.5178 ± 0.0002	0.5267 ± 0.0003	
7	0.5062 ± 0.0002	0.5151 ± 0.0002	0.5229 ± 0.0005	



Fig. 4. Ratios C(x) between the $(C_{2\pi}/A)_x$ values obtained for sources with nuclides incrusted at a depth *x* in the backing and the values of $(C_{2\pi}/A)_0$ for the same source but with the nuclides deposited on the backing surface.

Figure 4 also shows that, for a given incrustation depth in the source backing, the higher the alpha emission energy, the closer to unity is the ratio C(x), i.e., the higher the energy, the smaller would be the deviation in

the determination of the activity if the incrustation were not taken into account in the corrections to the counting rate. This is because the absorption of particles within the source support decreases with energy.

An extensive work by Berger [1] on the calibration of alpha-particle sources with a 2π geometry chamber also examined the effect of incrustation of radioactive material in the backing. Table 2 shows a comparison of our results for 3 MeV alpha particles with those in Berger's theoretical study for the three backings considered here. Only the results for 3 MeV can be compared, because the values corresponding to 5 and 7 MeV are not given accurately in Berger's work. One observes that our results do not precisely agree with Berger's, although the pattern of behaviour with energy and incrustation depth is similar.

In sum, one can conclude that if one does not include the incrustation of nuclides in the calibration of this type of source, but considers just deposition onto the backing surface, the deviations from the real value of the source's activity could be even greater than 1%. Therefore, the possibility of incrustation of radionuclides in alpha-particle sources is an important aspect to take into account in the standardization of alphaparticle emitters, for which the uncertainties associated with the activity must be strictly minimized.

Sources with nuclides uniformly distributed down to a depth \boldsymbol{x}

Finally, we simulated sources in which the alpha-particle emitting nuclides are assumed to be embedded uniformly in the backing, from the surface down to a given depth x. Figure 3 shows the spectrum of detected alpha particles in the particular case of 5 MeV and when the radionuclides are incrusted uniformly to a depth of 50 µg/cm² in a backing of Pt (233 Å). As observed, the low-energy tail associated with the 5 MeV alpha emission is somewhat less pronounced than in the case of incrustation to just a single depth of 50 µg/cm², also shown in the figure.

Figure 5 shows the ratios C(x) between the $(C_{2\pi}/A)_x$ values for these sources (nuclides uniformly distributed in the backing down to a depth x) and the values of $(C_{2\pi}/A)_0$ for sources with the nuclides deposited on the surface. As expected, for each backing, alpha-particle energy, and incrustation depth x, the values of C(x) are closer to unity than those obtained for the previous case of sources with nuclides incrusted to just a single depth x. This means that, in this case, the deviations produced in the determination of the activity if the incrustation were not considered would now be somewhat

Table 2. Values of the ratio $C(x) = (C_{2\pi}/A)_x/(C_{2\pi}/A)_0$ for alpha-particle sources of 3 MeV in which the radionuclides are incrusted at a single depth *x* within the backing. The uncertainties correspond to one standard deviation. Results reported by Berger [1] are given for comparison

Depth x (µg/cm ²)	Al		Ag		Pt	
	This work	Berger	This work	Berger	This work	Berger
10	0.9920 ± 0.0007	0.9915	0.9958 ± 0.0017	0.9936	0.9935 ± 0.0007	0.9940
20	0.9880 ± 0.0006	0.9861	0.9893 ± 0.0018	0.9894	0.9895 ± 0.0023	0.9908
30	0.9848 ± 0.0003	0.9816	0.9874 ± 0.0021	0.9858	0.9880 ± 0.0024	0.9882
40	0.9800 ± 0.0003	0.9775	0.9837 ± 0.0012	0.9825	0.9878 ± 0.0022	0.9859
50	0.9762 ± 0.0004	0.9738	0.9828 ± 0.0028	0.9794	0.9845 ± 0.0005	0.9838



Fig. 5. Ratios C(x) between the $(C_{2\pi}/A)_x$ values obtained for sources with nuclides incrusted uniformly from the surface down to a depth *x* in the backing and the values of $(C_{2\pi}/A)_0$ for the same source but with the nuclides deposited on the surface.

smaller. The pattern of behaviour of the C(x) values with energy and depth is, however, similar for the two types of source (with nuclides incrusted in the backing at a single depth x, and uniformly distributed down to a depth x in the backing).

Conclusions

We have studied the influence that the incrustation of nuclides in the source backing can have on the total detection efficiency in 2π sr measurements, and hence on the activity estimated for the source. A Monte Carlo

simulation procedure was used to model the behaviour of the alpha particles in the support material. The values of the total detection efficiency $C_{2\pi}/A$ were calculated for three backing materials (Al, Ag, and Pt) and for different alpha-particle energies, considering a wide range of incrustation depths in the source support. The results showed that, for the range of incrustation depths considered, the deviations produced in the determination of the activity if the incrustation of nuclides is not considered could be up to 2.4%. In addition, this work has shown that the Monte Carlo simulation code SRIM can be a good procedure with which to estimate the corrections needed in the measurement of alpha-particle sources with 2π detectors, especially when the radionuclides are embedded in the backing.

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