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Influence of source composition and particle energy on the determination of gross alpha activity

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HIGHLIGHTS

- SRIM code was applied to the study of gross alpha activity.
- SRIM validity was checked against experimental measurements.
- Influence of source composition and α-particle energy on the efficiency was studied.
- Influence of α-particle energy on the efficiency was studied.

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ABSTRACT

The influence of different source compositions and α-particle energies on the detection efficiency of a gas-flow proportional counter was examined using experimental measurements and Monte Carlo simulations. Efficiency variation with alpha-particle energy was very marked, being less significant with the substrate composition. These results show that the determination of gross alpha activity in an unknown sample must be carried out very carefully in order to give a correct estimation of its activity.
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1. Introduction

Gross alpha activity measurement (jointly with gross beta activity) is a technique extensively used in the initial screening of radioactivity in environmental samples. Since further and deeper analyses of such samples are usually far more expensive and laborious, laboratories generally first measure this parameter, and only continue with further analyses for those cases in which the gross alpha activity index of the sample exceeds some pre-set threshold.

There are different techniques for determining the gross alpha activity in a water sample, such as gas-flow proportional counting and liquid or solid scintillation (using ZnS(Ag) plates or plastic scintillators). The first type (gas-flow counting) is the most commonly used (Semkow and Parekh, 2001; Ruano Sánchez et al., 2013a, 2013b). There is now a broad set of standardized procedures which are recognized for the application of this technique (EPA, 1980; APHA, 2005; ISO, 2007). With some slight variations, all the procedures involve slowly evaporating an aliquot of the water sample on a circular striated stainless steel planchet using a heater. The source on the planchet is then measured in the gas-flow proportional counter. This same technique has also been used for the measurement of gross alpha activity in soil samples (Martín Sánchez et al., 2009).

With both soil and water samples, the main drawback is the self-absorption or energy loss of alpha-particles in the sample itself due to the thickness of the source substrate. Corrections must be applied to the experimental results to take these effects into account, together with the geometrical factor, in order to determine the detection efficiency.

To cover a wide range of thicknesses, an attenuation correction curve must previously be obtained from the measurements of standard sources prepared with different known thicknesses. Various works have dealt with finding line shapes that are appropriate for this curve. For instance, experimental data from the

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measurements of standards have been fitted to a quadratic (Suarez-Navarro et al., 2002), a power (Parsa et al., 2005), and exponential (Martín Sánchez et al., 2009) functions, among others. While the focus in these studies has been to determine the best fitting function, the influence of substrate composition or alpha-particle energy has not been analysed in any depth.

The aforementioned standardized methods in the experimental procedure suggest the use of different substrate compositions for the standard sources to determine the attenuation curve. The commonest substrate suggested is drinking water residue, whose level of radioactivity will be negligible compared to the quantities added artificially. Other works propose the use of salts commonly found in drinking water – sodium carbonate, sodium nitrate, or sodium chloride – but always taking any deviations related to the use of these salts to be unimportant (Baeza Espasa et al., 2012).

The recommended alpha-particle emitting nuclides to use in the fabrication of the standard sources are ^{209}Po, ^{241}Am, ^{230}Th, natural uranium, or some plutonium isotopes (Suarez-Navarro et al., 2002; Parsa et al., 2005; Martín Sánchez et al., 2009; Baeza Espasa et al., 2012; Ruano Sánchez et al., 2013a). This is because their alpha-particle emission energies are close to those of ^{226}Ra and natural uranium, which are the commonest radioactive contaminants found in environmental water samples.

The present work uses the SRIM Monte Carlo simulation code (Ziegler et al., 2010) to study the influence of the substrate composition and the standard radionuclide's alpha-particle emission energy on the attenuation curve representing the efficiency corrections versus source thickness. The simulation results were then checked against those reached experimentally when it was reasonably possible, measuring sources of known composition and several alpha-particle emitters with a gas-flow proportional counter.

2. Materials and methods

2.1. Gas-flow proportional counting

The experimental device used for the measurements was an MPC-9300 gas-flow proportional counter (Protean Instruments Corporation). This uses PR-10 gas (90% argon and 10% methane) for operation, and has an 80 μg/cm² Mylar sheet as the window entrance separating the source from the detector. The distance from the source backing to the detector is about 10 mm. Sources with up to 4.86 cm diameter can be measured. Simultaneous alpha and beta activity counts are usually determined with a 1605 V bias voltage. If only the determination of gross alpha activity is required, a voltage of 915 V can be applied, as recommended by the manufacturer. The alpha-particle detection efficiency values obtained using these two detection modes were found to be equivalent, so that only single alpha-particles detection mode was used for the present work.

The method of preparing standard sources for measurement was as follows. A small amount of either sodium nitrate ($NaNO_3$) or sodium carbonate (Na_2CO_3) was placed on an 18.6 cm² stainless steel counting planchet. A small volume of de-ionized water was added, followed by stirring until the salt was completely dissolved. A known activity of the radionuclide of interest was added, followed by stirring and evaporation to constant weight (Ruano Sánchez et al., 2013a; Martín Sánchez et al., 2009). Some sources showed a clear visual inhomogeneity due to clustering formed in the evaporation process and they were discarded. ^{241}Am was used with both substrates, while ^{209}Po was used only with the $NaNO_3$ substrate.

2.2. Monte Carlo simulations

SRIM (Stopping and Range of Ions in Matter) is a well-known Monte Carlo simulation program (Ziegler et al., 2010), which has

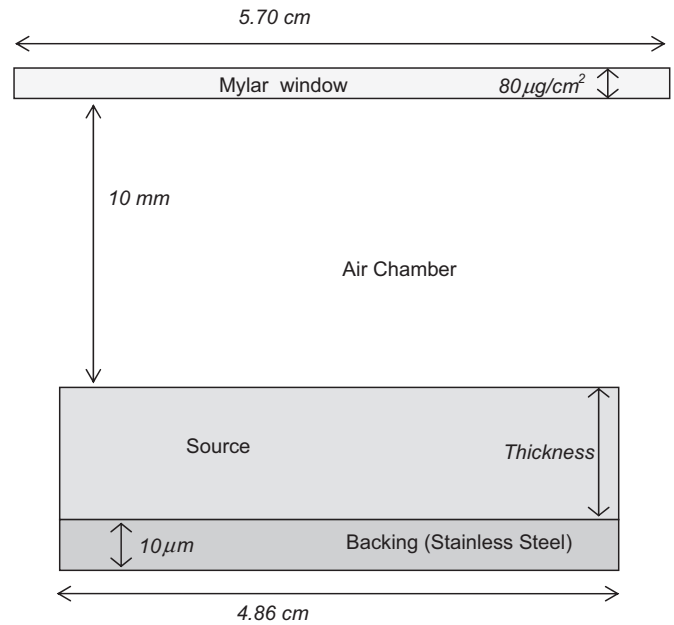

Fig. 1. Scheme of the experimental array considered for the simulation process.

been extensively used to study interactions of heavy charged particles, and tested in many applications, including simulations of the interactions of alpha particles in matter (Jurado Vargas and Fernández Timón, 2004; Fernández Timón et al., 2010; Jurado Vargas and Fernández Timón, 2012). We applied it here to study the alpha particles detected with a gas-flow proportional counter. Fig. 1 shows the scheme of the experimental set-up considered for the simulations.

The simulation time for each individual simulation was of the order of 2 h on a 2.33 GHz Intel Core2 Duo PC. The input files contained the initial characteristics of the incident alpha particles (energy, direction, and position in the source), and the composition of the source. Data related to the detector system (air gap between source and detector, Mylar window thickness, etc.) were put into the SRIM main menu. A text file (TRANSMIT.TXT) stored the characteristics of the alpha particles having passed the detector window, and which were then assumed to subsequently be detected. For each simulation, the ratio between the number of detected particles and the initial number of simulated particles gave the system's detection efficiency. Several simulations were performed using the same value of all the parameters but with different random number seeds to estimate the uncertainty related with each efficiency value. Only this stochastic contribution was included in the uncertainty estimates because simulation is generally unable to consider the uncertainty associated with the physical parameters involved in the interaction processes.

3. Results and discussion

3.1. Comparison of the simulation results with experiment

Monte Carlo simulations were performed with the two source substrates used experimentally ($NaNO_3$ and Na_2CO_3), considering source thicknesses from 1 to 14 mg/cm². Two alpha-particle energies were considered for this first series of simulations: 5.486 MeV, principal emission of ^{241}Am, and 4.880 MeV, corresponding to the ^{209}Po emission. Four simulations, each including 5×10^4 alpha particles, were performed for each individual case.

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