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An advanced method of activity determination of large area beta emitting sources

Andrej Javorník, Anton Švec *

Slovak Institute of Metrology, Karloveská 63, 842 55 Bratislava, Slovak Republic

HIGHLIGHTS

- Efficiency tracing transmission method of beta activity determination.
- Efficiency determined by means of a parameter independent of initial absorption conditions.
- Parameter is derived from two counting results obtained with using a test foil.
- Particularly useful for calibration and measurement of radionuclide standard sources.

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ABSTRACT

The presented advanced method of activity determination of large area beta emitting sources is based on a version of efficiency tracing method using a test foil placed between the source and a conventional large area detector. It is shown that the total efficiency of the measuring system may depend on a dimensionless parameter derived from the difference in count rates caused by inserting the test foil while other disturbing effects are mostly reduced or compensated.

1. Introduction

Large area sources used for calibrations of area activity measuring instruments are specified by ISO 7503-1 (ISO, 1988), ISO 8769 (ISO, 2010) and IEC 60325 (IEC, 2002) standards. These sources shall be accompanied by a certificate containing both the surface emission rate and the activity. These quantities are mutually related by the source efficiency which depends on the source self absorption. There is a general opinion that the activity value is inaccessible to direct measurement and the instrument calibration is performed on the surface emission basis (ISO, 2010). According to the ISO 7503-1 methodology (ISO, 1988) the total efficiency of a surface contamination measurement should be separated into source and detector efficiencies where the detector efficiency is determined as a ratio of the detector response to the surface emission rate of the calibration source. High self-absorption in some sources causes their efficiencies are low and the relationship between their surface emission rate and activity is regarded as inferior. No wonder that the instrument-related standard (IEC, 2002) ranks these meters into ±25% class. The revised ISO standard 8769 (ISO, 2010) contains no mention of the source efficiency and is limited to the requirement to state the source activity with rather high standard uncertainty (±10%) together with much more accurate surface emission rate. It makes the situation no easier: the aim to measure surface contamination is to evaluate activity per unit area and the fact that the instrument calibration factor based on the emission rate is much less dependent on the source construction does not improve the total efficiency required for the activity determination. However, the source self absorption causes also specific changes in the spectral characteristics of the emitted radiation that can be utilized for measured data corrections. Emitted radiation itself carries information about absorption effects which passed and additional measurements of a kind may reveal them. Such an approach differs from conventional standard techniques and brings some advantages.

Recently we have tried to develop a direct method for activity determination of large area sources (Švec et al., 2006) that followed similar experiments of Janßen and Klein (1996). However, it was found soon that common large area proportional or scintillation detectors of beta radiations are never large enough in comparison with the beta-particle ranges in air which absorption properties are rather weak, especially for harder beta radiations and require corrections for barometric pressure variations. Moreover, changes of absorption conditions by varying the air layer thickness are always accompanied with solid angle changes. In the

* Corresponding author. Tel.: +421 260 294 657; fax: +421 260 294 670.
E-mail address: svec@smu.gov.sk (A. Švec).

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present study, it was therefore decided to keep geometrical conditions fixed and to vary absorption conditions by using suitable solid test foils which alter just one parameter—the overall absorption of particles.

2. Theoretical

Absorption of beta particles is often described (Baltakmens, 1970) by exponential curves similar to the attenuation law of gamma rays. The absorption coefficient obtained by regression analysis was also presented like a characteristic parameter for given absorbing material which correlates with the maximum or mean energy of beta radiations (Baltakmens, 1977). In our initial work (Švec et al., 2006) an experimental parameter r equal to an effective mean free path for a given radiation energy in given absorbing material has been used for testing the absorption relationship. This parameter corresponds to reciprocal value of the absorption coefficient and would be constant only in the case of pure exponential law but becomes dependent on absorption conditions in all other non-exponential cases. More detailed numerical experiments published by Berger (1998) for 19 selected radionuclides embedded in an aluminum pad in various depths up to 10 μm also showed that the real absorption of beta radiation differs from the exponential approximation and there is some correlation with the level of absorption expressed e.g. by thickness of the absorption layer already passed. For practical purposes he further proposed to find the half-value thickness of mylar foils used as an auxiliary absorbing material covering the sources. This parameter, equivalent to the absorption coefficient, was related to sources efficiencies. More recent data published by Stanga et al. (2011) for five most common radionuclides are fully consistent with those of Berger. Moreover, Stanga et al. proposed an empirical three-parameter analytical curve which fitted the numerical data very well in the range from virtual zero to 20 μm thick absorption layers of aluminum. Adopting their notation, the effective parameter r introduced by Švec et al. (2006) can be related to the analytical function of Stanga et al. (2011) by following transformation

$$r(x) = -\frac{I}{dI/dx} = -\frac{\varepsilon}{d\varepsilon/dx} = \frac{1}{b + (c/(2\sqrt{x}))} \tag{1}$$

where I is the net count rate registered after passing some absorption layer(s) of thickness x and $\varepsilon = I/A$ is total efficiency of the measurement while parameters b and c are coefficients of Stanga's empirical curve. It should be noted that $r(x)$ is only two-parameter unambiguous function of the thickness x since the parameter a characterizing the zero-thickness efficiency has been eliminated by transformation (1). Now, the $\varepsilon(x)$ function, whose values are difficult to obtain experimentally due to insufficient knowledge of partial and total values of x, can be substituted by another empirical function $\varepsilon(r)$ representing the same relationship with even fewer parameters. In our former work we treated $\varepsilon(r)$ as an unspecified continuous function approximated by a parabolic curve which shape and position were determined by a numerical procedure equivalent to LSQ regression analysis in each of seven experimental points separated by narrow equidistant intervals of x. Such an approach particularly suited experiments using air as the absorbing medium because the air layer thickness can be easily adjusted in sufficiently small steps of about 0.1 mg cm^{-2}. Yet, our experimental results (Švec et al., 2006) cannot be directly compared with data newly published by Stanga et al. (2011) because of differences in absorbing materials (air vs. aluminum), radionuclides and necessary changes in the solid angle. However, the shape of the curves calculated by Eq. (1) and their relative positions confirmed the feasibility to obtain the $\varepsilon(r)$ relationship experimentally.

Following considerations concern ways to improve and simplify all the procedure of experimental data acquisition and processing. First, as discussed above, the method of changing the air absorption layer thickness by a movable table has been replaced by a more comfortable method of insertion thin test foils into a narrow slit between the source and the detector while keeping the distance between the source and detector fixed. When a selected test foil of a suitable material with thickness Δx is inserted in the slit, its presence causes a change ΔI in the registered count rate which is further used to determine the counting efficiency from an efficiency curve. For this purpose, the derivation in the relationship (1) has been replaced by differentiation as follows:

$$r(\Delta x) = -\frac{I_x - I_0}{\Delta I/\Delta x} = \frac{I_x - I_0}{I_x - I_{x+\Delta x}}\Delta x \tag{2}$$

where I_x and $I_{x+\Delta x}$ are registered count rates without and with the test foil inserted and I_0 is the background count rate. Like in Eq. (1), the minus sign respects the negative correlation between Δx and ΔI (increasing x causes decreasing I). It can be expected that characteristics of this new differential relationship and the former one based on continuous derivation (1) will be very similar.

Another consideration led to changing the form of $\varepsilon(r)$ display. Since the foil thickness Δx in Eq. (2) acts as an experimental constant it can be omitted if always the same test foil is used. So the basic model of the proposed experiment with a foil of thickness Δx has been reconsidered, defining a dimensionless parameter $p = r/\Delta x = I_x - I_0/I_x - I_{x+\Delta x}$ in the form

$$\varepsilon(p) = \varepsilon\left(\frac{I_x - I_0}{I_x - I_{x+\Delta x}}\right) \tag{3}$$

This relationship can be obtained experimentally and afterwards used as a calibration curve for testing sources using always the same test foil characterized by its material and thickness Δx. The dimensionless parameter p can be regarded as a figure of merit related to various absorption conditions represented by variations of x and characteristic for a given radionuclide at given experimental arrangement with the selected test foil. Then the relationship (3) is a characteristic function for this type of experiments which relates experimentally available data and the counting efficiency.

3. Experimental

Our experimental work was performed using a common household aluminum foil like the test foil. Its area density of 2.54 mg cm^{-2} was determined by weighing and corresponded to about 9.5 μm thickness. Other foils prepared from 0.37 mg cm^{-2} metalized mylar were used for simulation of absorption differences in active and protective layers of the source while aluminum foils were used in the case of harder radiations to increase the absorption effect. A plastic scintillation detector sized 20 cm × 30 cm with a mylar window of 0.88 mg cm^{-2} area density (an aluminum equivalent about 3.8 mg cm^{-2} including air dead layers) was connected to a multichannel analyzer operated in the integral mode which served as a single wide-channel counter in the energy range from about 6 keV (set by the low level discrimination control) up. Certified standard area sources, of ^{14}C, ^{36}Cl, ^{60}Co, ^{90}Sr+Y, ^{147}Pm and ^{204}Tl, 11 cm × 15 cm traceable to CMI, were placed close to the detector input window leaving about 5 mm wide slot for inserting the foils. Except the common background I_0, two count rates were registered each time: one I_x with the bare source and another one $I_{x+\Delta x}$ with the test foil inserted. Preset live time were 10^3–10^4 s and 5×10^5–5×10^6 counts registered every time. The efficiency ε=(I_x–I_0)/A was calculated from registered count rates using the certified activity A of the source and associated to the p=(I_x–I_0)/(I_x–$I_{x+\Delta x}$) ratio. Then the source was gradually covered by one or several additional absorbing mylar or aluminum foils and the counting procedure was repeated using always the same aluminum