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Experimental determination of the self-absorption factor for MTR plates by passive gamma spectrometric measurement

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ABSTRACT

The measurement of the absolute activity or the mass of radioactive substances by gamma spectrometry needs to include a correction for the radiation absorption inside the source volume, the so-called self-absorption factor. It depends on geometry and material composition of the source, the detector geometry and on the geometrical arrangement of source and gamma radiation detector; it can be calculated if full information about all that is available. This article however describes how to determine the self-absorption factor from measurements if the radiation sources are plates of uranium fuel with typical parameters of nuclear fuel for MTR reactors and without using detail information on the source geometry, thus allowing easy inspection without relying on – potentially falsified – declarations on the internal properties of the fuel objects and without calculation.

1. Introduction

Materiel testing reactors (MTR) are normally fed with uranium, which is contained in aluminium fuel plates. The uranium enrichment is often in the order of magnitude of 20% or 90%. The mass of the fissile isotope ^{235}U has to be declared to the International Atomic Energy Agency (IAEA) or to EURATOM and is then subject to inspection measurements of nuclear safeguards inspectors. They have to verify the ^{235}U mass (or the mass of the element U and its enrichment), independent on information from the owner. Typically, this nuclear fuel has the form of rectangular Al plates with less than 0.2 cm thickness and an active, uranium containing area up to about 60 cm × 7 cm. The active area is covered on both sides with an Al cladding of about 0.04 cm thickness, whereas the central volume is filled with a matrix of uranium oxide and aluminium or of UAl_3 and aluminium. The uranium mass per unit area varies in a range from about 0.02 to 0.4 g U/cm^2 of the active plate region, the ^{235}U mass per unit area from 0.02 to 0.22 g ^{235}U/cm^2. The self-absorption factor for perpendicular emission varies between 0.73 and 0.97 for the 186 keV line. Intermediate products in fuel factories can have even higher U masses per cm^2 and consequently smaller self-absorption factors.

One technically implemented approach for the ^{235}U mass determination is as follows [1]: it is assumed, that the active area can be described reasonably well as a homogeneous rectangle. The emission rate of 186 keV photons of ^{235}U is measured with a gamma scan along the fuel plate. In the same scan, a transmission measurement is made using a ^{75}Se point source, which emits photons at the energies 135.9 and 264.5 keV. Since the attenuation coefficients of U and Al are sufficiently different at these energies it is possible to determine the mass per unit area for these two elements. With this knowledge, the self-absorption for the 186 keV can be determined. The measured emission rate, corrected with the self-absorption factor, delivers finally the ^{235}U mass. Unfortunately, the use of the radioactive transmission source causes practical problems; it also needs to be replaced often due to the rather short time of half life of only 120 days.

The purpose of the present paper is to propose and to validate experimentally a new approach for determining the self-absorption in a MTR plate.

2. Principle of the methods

The count rate CR_β for photons with an energy E emitted from an isotope j in a plane uranium fuel plate and measured by a point detector under an observation angle β see Fig. 1(b) is described by

$$CR_\beta = m_u \times enr \times N \times \varepsilon \times \frac{1}{A_j} \times \frac{1}{t} \times \lambda_j \times I_{\gamma,E} \times SG_{tot}, \quad (1)$$

with m_u is the total mass of uranium in the slab, enr the ^{235}U enrichment, N the Avogadro number, ε the intrinsic detector efficiency, A_j the atomic weight of isotope j, t the counting time, λ_j the decay constant of ^{235}U, μ the mass attenuation coefficient in

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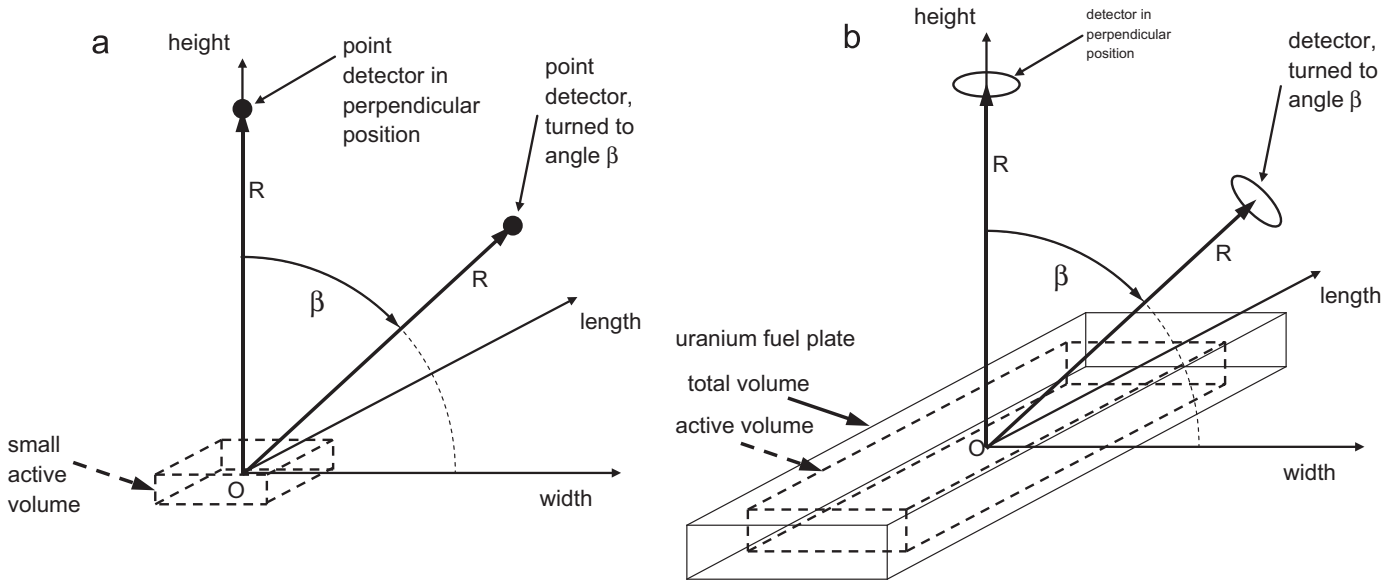

Fig. 1. Detector positions and angle β in relation to a small uranium plate (a) and a real, extended uranium fuel plate (b).

the slab, ρ the density in the slab, x_i, y_i, z_i the coordinate of the intercept point I where the photon leaves the plate, $I_{\gamma,E}$ the gamma branching ratio, SG_{tot} is the integral over the source volume.

$$SG_{tot} = \iiint \frac{\exp(-\mu \times \rho\sqrt{(x-x_i)^2+(y-y_i)^2+(z-z_i)^2})}{x^2+y^2+(z-R)^2}\,dx\,dy\,dz. \quad (2)$$

It describes both the geometry and the internal absorption. x_i, y_i and z_i depend on x, y and z, respectively, and on position and shape of the detector. Under certain geometrical conditions which will be discussed further down, the integral may approximately be replaced by two factors, the self-absorption S_β and a geometry factor G, which is practically independent on β

$$SG_{tot} = S_\beta \times G. \quad (3)$$

The two factors can be calculated. For the calculation of S_β, the exact internal material composition has to be known. If, as in our case, the amount of uranium in the plate has to be measured, it is unknown and S_β cannot be calculated. This article describes an approach for the determination of the self-absorption factor S_β by measurement.

2.1. Principle

The principle of the method is to measure the emission of photons from two angles (Fig. 1(a) and (b)), for instance the intense radiation at 186 keV. The two emission rates are different due to different self-absorption factors. The ratio of the two emission rates is systematically related to the self-absorption; hence it can be used to determine the two self-absorption factors for the two directions, which will finally be applied to determine the ^{235}U mass.

The principle of this approach can be explained best if we make – for this explanation – the following assumptions (Fig 1(a)):

1. The detector is a point detector.
2. The fuel object is a plane disc.
3. Its lateral extensions are small in comparison to the distance between source and detector. For the moment we imagine also that the longitudinal extension is small in comparison to the distance from source to detector (Fig. 1(a) and (b)). This limitation is not necessary and further down is explained why.
4. Its thickness is negligible in comparison to its lateral extension.
5. The Al cladding thickness is always known, hence the absorption in this layer can be corrected for; the values are all very close to 0.04 cm.
6. The uranium mass per unit area is smaller than about half of the "infinite thickness" for enrichment measurements using the enrichment metre principle [2], i.e. below 3 g U/cm^2.

Under the above conditions the emission rate in perpendicular direction is proportional to the self-absorption S_0 factor for the U–Al-matrix

$$S_0 = \frac{(1-e^{-\mu\rho d})}{\mu\rho d}, \quad (4)$$

where d is the thickness of the slab.

If we now turn the detector by an angle β out of its perpendicular position but keep it in a constant radial distance R from the centre of the source (Fig. 1(a)), we get an emission rate proportional to the self-absorption factor S_β

$$S_\beta = \frac{(1-e^{-\mu\rho d/\cos(\beta)})}{\mu\rho d/\cos(\beta)}. \quad (5)$$

The ratio $R_{\beta,0}$ of the two emission rates is

$$R_{\beta,0} = \frac{S_\beta}{S_0} = \frac{(1-e^{-\mu\rho d/\cos(\beta)})}{(1-e^{-\mu\rho d})/\cos(\beta)}. \quad (6)$$

The relation between the ratio $R_{\beta,0}$ and the self-absorption S_β for the angles $\beta=0^\circ$ and 60° is shown in Fig. 2. Along the curves, the parameter $\mu\rho$d, changes. Since it does not play any role which one of the three factors of the product changes, the curves are independent on the value of μ and they can be used for all gamma radiation peaks of ^{235}U at any energy E. The ratio $R_{\beta,0}$ is equal to the ratio of the count rates CR_β/CR_0 since all factors in Eqs. (1) and (3) except β are identical. The ratio CR_β/CR_0 can be measured. Using the curves of Fig. 2, the not measurable self-attenuation factors S_0 and S_β can be derived, hence we can experimentally determine the self-absorption factors. For the angle $\beta=60^\circ$ and thin plates, we have even a direct proportionality between the