Density measurement by radiometric method with gamma irradiation from sources of low activity

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Abstract. The method to obtain the density of the media by gamma-irradiation is proposed. The main issue of this method is the analytical calculation of the spectrum measured by gamma-spectrometer; this calculation provides information about scattered gamma-quants in a beam. Thus, this method precludes the use of collimators in experiments, therefore, this made it possible to avoid using very high-activity sources of ionizing radiation: we present the experimental results with gamma-sources of activity ~10⁴ Bq.

1. Introduction

Some physical parameters can be determined remotely by the use of the gamma-irradiation of the samples [1–9]. Sometimes such methods are referred as ‘X-ray CT (computed tomography) methods’, or as ‘gamma-methods’; we will use the term ‘radiometric methods’. We could say that idea of such methods is old and clear, but, in our opinion, it is more correct to say that this idea is simple and obvious.

The main feature of the radiometric methods is the measurement of the physical density: the density of the given substance can be obtained from the expression for the attenuation of the flux of gamma-quants:

\[ J = J_0 \exp(-\mu_m \rho x) \]  

(1)

from which we have

\[ \rho = \frac{1}{\mu_m x} \ln \left( \frac{J_0}{J} \right). \]  

(2)

Thus, determining mass attenuation coefficient \( \mu_m \) from reference data (for example, from the NIST database [10]) and obtaining \( x \) (width of the sample), \( J_0, J \) (fluxes of gamma radiation without/with substance) from experiment, we can calculate the density \( \rho \). Many parameters so could be obtained, such as a composition of a mixture, a concentration of a solution, a void fraction in a liquid etc. Different goals lead to different requirements for the source of ionized radiation.

For instance, the mass concentration of the solution \( C \) can be obtained by measuring linear attenuation coefficient...
\[ \mu = \mu_m(C)\rho(C) = \frac{1}{x} \ln \frac{J_0}{J}. \]  

(3)

Hear the mass attenuation coefficient of the water solution is

\[ \mu_m(C) = C \mu_m^{\text{dis}} + (1 - C) \mu_m^{\text{wat}}, \]  

(4)

where \( \mu_m^{\text{dis}} \) is the mass attenuation coefficient of the dissolved substance, and \( \mu_m^{\text{wat}} \) is the mass attenuation coefficient of the water. Measuring parameter \( \mu = \frac{1}{x} \ln \frac{J_0}{J} \), one can obtain \( C \) from the Eq. (3), but additional function \( \rho(C) \) is needed. To obtain good result for \( C \) (i.e. the low indeterminacy of its value) one needs the source of gamma-quants of the quite low energy \( E_\gamma \) : if the Compton scattering is the dominant effect of the attenuation of gamma-ray, then mass attenuation coefficients of the different elements are close to one another (except of hydrogen and heavy elements). But coefficient \( \mu_m^{\text{dis}} \) must be different from \( \mu_m^{\text{wat}} \) significantly, that is why the energy of the gamma-quants for these measurements must be sufficiently low (the dominant effect of the attenuation of gamma-ray should be photoeffect).

We have another situation for the measurement of the density of the solution. In this case, conversely, we can use the source of the high-energy gamma-quants: if \( \mu_m^{\text{dis}} \approx \mu_m^{\text{wat}} \), then the dependence \( \mu_m(C) \) can be safely neglected, so one can obtain the density of the solution of the unknown concentration.

But, anyway, equation (1) is correct only for a narrow beam of a gamma-radiation. For the wide beam of gamma-rays the scattered gamma-quants appear, so the collimators must be used to cut these additional quants. Because of this reason, radiometric methods demand high-activity gamma-sources: for example, in [5] activity of isotope \(^{137}\text{Cs}\) – source of gamma-radiation – was \(2.4 \times 10^{11}\) Bq.

Indeed, for the gamma-source of the activity \( A \) and the quantum yield \( n \) we have the equation for the number of counts per second in detector

\[ N = An \frac{r^2}{4l^2}, \]  

(5)

where \( r \) is the radius of the collimator, \( l \) is the distance between the gamma-source and the collimator. Usually \( l \approx 1 \) m and \( r \approx 1 \) cm; considering a fact that \( N \) must be much greater than background counting rate (~1 s\(^{-1}\)), i.e. \( N \) should be \( \approx 10^2 \) s\(^{-1}\), we can see from Eq. (5) that \( A \) must be not less than \( \approx 10^7 \) Bq. Considering attenuation of the radiation in the sample, one may enlarge this estimation of the activity of the gamma-source on a magnitude.

Thus, radiometric measurements can be dangerous for a health of researchers, and other methods to take account of the scatter radiation in the experiment would be developed. In our work we propose the radiometric method for radioactive sources of a low activity: scatter radiation can be subtracted by means of the mathematical and physical methods, not only by the technical one. Therefore, it is possible to avoid using collimators in the experimental scheme, and the total amount of gamma-quants would be defined only by the size of our detector, not by the aperture of the collimator. Therefore, the activity of the gamma-source can be low.

2. **Theoretical background**

Scintillation detectors allow one to measure a spectrum of gamma-quants, but the picture of this spectrum is rather ‘washed’: these spectrometers have finite (and quite large) resolution, so we would see distribution – i.e. artificial distribution – on energy of gamma-quants instead of the \( \delta \)-function even
for the monoenergetic source (source of the gamma-quants at the single energy). Fortunately, the form of this artificial distribution can be easily predicted.

For many reasons, the absorption peak (AP, peak, registered by the scintillation spectrometer) of the non-scattered radiation from the monoenergetic gamma-source must have the Gaussian form:

$$I^{(NS)}(\varepsilon) = \frac{B}{\sqrt{2\pi D}} \exp\left(-\frac{(\varepsilon - \varepsilon_0)^2}{2D}\right),$$  \hspace{1cm} (6)

where $\varepsilon_0$ is the energy of the gamma-quants emitted by the gamma-source, $B$ is the total intensity of this peak, $D = \sigma^2$ is the dispersion of the given gamma-spectrometer. Thus, FWHM (full width at half maximum) parameter $\Delta$ of the gamma-spectrometer at the given energy of gamma-quants is

$$\Delta = \sigma \sqrt{2\ln 2}.$$  \hspace{1cm} (7)

For our MKS-15EC scintillation spectrometer at peak of 662 keV (from $^{137}$Cs) we have $\sigma = 25$ keV, $\Delta = 59$ keV, thus, the resolution of the spectrometer for energy 662 keV is $\Delta/\varepsilon_0 = 9\%$.

As we can see from Fig. 1, the formula (6) provides correct interpretation of the measured AP. Note that this gamma-spectrum was obtained from $^{137}$Cs gamma-source with empty container (for the investigated substance); so this spectrum holds the negligible presence of the scattered radiation from the walls of this container (as well as the scattering in spectrometer).

![Fig 1](image.png)

**Figure 1.** The real form of the absorption peak (1) measured by spectrometer MKS-15EC and the Gaussian curve (2).

However, for the scattered radiation (scattered in the investigated substance) AP has non-Gaussian form. To obtain intensity of the non-scattered radiation (i.e. $J$ for equation (2)) we must reduce total intensity of the AP by subtracting the scattered gamma-quants.

Considering a fact that we are interested only in gamma-quants with energies $\varepsilon \sim \varepsilon_0$ (note that scattered gamma-quants have lesser energies, i.e. $\varepsilon < \varepsilon_0$), we can represent cross-section of the Compton scattering [11] (the Klein–Nishina–Tamm formula) in the media.
\[ \eta(\varepsilon) = \frac{d\phi}{d\varepsilon} = C_0 \left[ \frac{\varepsilon_0}{\varepsilon} + \frac{\varepsilon}{\varepsilon_0} + \left( \frac{\mu}{\varepsilon} - \frac{\mu}{\varepsilon_0} \right)^2 - 2\mu \left( \frac{1}{\varepsilon} - \frac{1}{\varepsilon_0} \right) \right] \] (8)

in linearized form:

\[ \eta(\varepsilon) = C \left( 1 + \frac{\mu(\varepsilon - \varepsilon_0)}{\varepsilon_0^2} \right), \] (9)

where constants \( C_0 \) and \( C = 2C_0 \) depend on the total amount of the scattered gamma-quants (which depends on the thickness of media) and can be found from experimental data; \( \mu = 511 \text{ keV} \) is the rest mass of the electron, and \( \frac{\varepsilon_0\mu}{\mu + 2\varepsilon_0} \leq \varepsilon \leq \varepsilon_0 \).

Figure 2. Experimental form of the gamma-spectrum near the absorption peak of \(^{137}\text{Cs} \) (1) and its theoretical approximation (2) by Eq. (12); curve (3) is the contribution of the scattering with linearized cross-section (Eq. 11).

Using (9) and considering the Gaussian form for every peak of the scattered quants, we can obtain the ‘scattered-part’ in AP in form

\[ I^{(S)}(\varepsilon) = \int_{\varepsilon_m}^{\varepsilon_0} C \left( 1 + \frac{\mu(\varepsilon' - \varepsilon_0)}{\varepsilon_0^2} \right) \frac{1}{\sqrt{2\piD}} \exp \left( - \frac{(\varepsilon - \varepsilon')^2}{2D} \right) d\varepsilon'. \] (10)

On the one hand, the lower limit of this integral must provide correctness of the linearization: \( \frac{\varepsilon_0 - \varepsilon_m}{\varepsilon_0} \ll 1 \): integral (10) lost any sense at the left branch of the cross-section, where function \( \eta(\varepsilon) \) grows with decrease of energy \( \varepsilon \). From another point of view, \( \varepsilon_m \) must be sufficiently small,
because the result – the form of the AP with the center at $\varepsilon_0$ – cannot depend on such parameter. The value $\varepsilon_m = 500$ keV satisfies both conditions.

From (10) we have:

$$I^{(S)}(\varepsilon) = \frac{C}{2} \left( 1 + \frac{\mu (\varepsilon - \varepsilon_0)}{\varepsilon_0^2} \right) \left[ \text{erf} \left( \frac{\varepsilon - \varepsilon_0}{\sqrt{2D}} \right) - \text{erf} \left( \frac{\varepsilon_m - \varepsilon}{\sqrt{2D}} \right) \right] +$$

$$+ C \frac{\mu}{\varepsilon_0^2} \sqrt{\frac{D}{2\pi}} \left[ \exp \left( - \frac{(\varepsilon_m - \varepsilon)^2}{2D} \right) - \exp \left( - \frac{(\varepsilon - \varepsilon_0)^2}{2D} \right) \right], \quad (11)$$

where $\text{erf}(x) = \frac{2}{\sqrt{\pi}} \int_0^x e^{-y^2} dy$ is the error function.

Thus, the total form of AP is the sum of (6) and (11):

$$I^{(AP)}(\varepsilon) = I^{(NS)}(\varepsilon) + I^{(S)}(\varepsilon). \quad (12)$$

As one can see on Fig. 2, total form of the AP can be represented sufficiently well by Eq. (12). Note that we use value $\varepsilon_m = 500$ keV, thus, the left part of the calculated spectrum on Fig. 2 has artificial diminution at energies $\varepsilon \sim 500$ keV; this fact has no influence on the form of the AP.

3. Experiment
To test our method we investigated the density of the corundum powder Al$_2$O$_3$ (electrocorundum; density $1.048 \pm 0.018$ g/cm$^3$ was determined gravimetrically) with the isotope $^{137}$Cs (activity is $1.7 \times 10^4$ Bq) as a source of gamma-radiation. Gamma-spectrum was collected with NaI(Tl)-scintillation detector MKS-15EC.

Experimental spectrum and its reconstruction (with parameters $B=375$ in Eq. (6) and $C=55$ in Eq. (11)) are presented on Fig. 2. The density of the corundum powder can be obtained from Eq. (2) and Eq. (12) by two ways:

1) using analytical function (6) for non-scattered radiation, i.e. in Eq. (2) we have $J = \int I^{(NS)} d\varepsilon$;

2) taking integral for experimental data $\int I^{(EXP)} d\varepsilon$ numerically and subtracting contribution of scattered quants in form of (11), i.e. in Eq. (2) we have $J = \int I^{(EXP)} d\varepsilon - \int I^{(S)} d\varepsilon$.

The first way is preferred because in this case we have simple ratio of constants $B$ from Eq. (6) (for intensity of the non-scattered gamma radiation with/without investigated substance) instead of $J_0 / J$ in Eq. (2), i.e. $J_0 / J = B_0 / B$.

For $\mu_m = 0.0758$ cm$^2$/g [10] we have from Eq. (2) $\rho = 1.013$ g/cm$^3$. Uncertainty of the value of $\delta\rho$ can be calculated through error of parameters $J$ and $J_0$, which can be found from experimental data for collected gamma-spectra by statistical methods. Thus, $\delta\rho$ is rather large: $\sim 3–5\%$, because of the imperfection of the spectra measured by our spectrometer (see Fig. 1 and Fig. 2, where this imperfection is clear).

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4. Conclusion
Equation (1) is correct only for the narrow beam of the non-scattered radiation, but we always have a wide beam of gamma-quants under conditions of real experiment. Thus, we have two choices:

1) To cut scattered quants by collimators. This is a direct method; it is widely used, but the activity of the source of ionized radiation during such measurements must be very high. Thus experiment becomes complicated because the system of the radiation protection becomes an essential part of the experimental setup; this study is potentially dangerous for the health.

2) To exclude scattered quants from the consideration theoretically by the use of mathematical equations for the well-known physical processes: the scattering of gamma-radiation in the media is the well-understood process, so we can obtain the total amount of the scattered gamma-quants through the shape of the measured gamma-spectrum. In this case we can use in experiment the wide beam of quants from the low-activity gamma-source.

As we can see from Fig. 2, the gamma-spectrum can be approximated by Eq. (12) sufficiently good. This fact allows us to determine the scattered gamma-quants analytically, thus the need for collimators in experiment was avoided, and the gamma-sources of the low activity were used in our researches: activity of the isotope $^{137}\text{Cs}$ was 17 kBq.

In compare with [5–7], we used the gamma-source with the activity at seven magnitudes weaker, but achieved quite suitable result for the density of the investigated media. Uncertainty of the results was rather large (a few percent), but we have no doubt that it is possible to improve the measurements by using an appropriate gamma-spectrometer.

In fine, we must note that for the large width of the sample ($x$ in Eq. (1)) one must take into account multi-scattered quants, so this theory must be developed further.

5. References
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