A Method for the Estimation of the Weak Gamma-Activity Distributed in the Thick Medium

By

M. ISHIYAMA*, Y. NAGAHARA*, M. OKANO**,
M. TAKANO* and M. TAKEUCHI*

(Received July 2, 1962)

Abstract

A simpler method for the estimation of the gamma-activity distributed in the thick medium is presented. This method was proved to be useful by the exemplifying experiment where the potassium content in sand was obtained with rather good accuracy.

Introduction

When the source intensity per unit volume is weak and also the extraction of the radioactive material in object is difficult or undesirable from some other reason, it is neccessary to take a large amount of the source material, in order to get the meaningful countings. Thus, in these cases, the self-absorption and the self-scattering of the radiation (hereinafter will be called simply as self-absorption) by the source material itself, and both the geometrical and the intrinsic efficiency of the detector used, would present rather complicated problems. Especially, the self-absorption of the gamma-ray is strongly dependent upon the physical properties of the source material, the geometrical shape of the source and the gamma-ray energy. So, it is difficult to give an exact definition to the "self-absorption coefficient". Moreover, even if the definition were given, it would be powerless in the practical application.

According to this method, the effects of the self-absorption need be neither measured nor estimated from other independent measurements or calculations. The intensity of the gamma-ray emitter can be obtained from a series of measurements by treating the effects of the self-absorption parametrically.

* Physics Department, Yokohama National University, Yokohama.
** Radiation Laboratory, Institute of Physical and Chemical Research, Tokyo.

Principle of the Method

Now, refering to Fig 1 and 2, the principle of this method will be described. For simplicity, the detector and the source container are assumed to be cylindrical, though they may be of arbitrary shape.

Using the following notations;

n—disintegration rate of the radioactive material, which is distributed uniformly in the source material, per unit volume of the source material.

$G(x)$—total (or, photopeak) efficiency of the detector (including both the geometrical and intrinsic efficiencies) for the gamma-ray emitted from the radioactive material in the region between x and $x+dx$.

$F(x)$—attenuation factor for the gamma-ray (emitted at height x) by the source material between o and x.

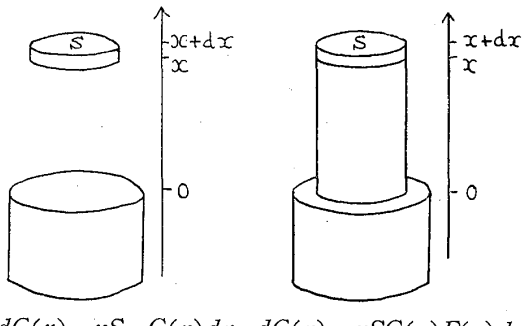
S—cross-sectional area of the container.

we can express the counting rate $C(h)$, where the container is filled up to height h with the source material, as follows;

$$C(h)=nS\int_0^h G(x)\cdot F(x)dx \tag{1}$$

From the definitions, it is obvious that $F(x)$ must satisfy the boundary conditions;

$$F(x)=\begin{cases}1 & \text{at} \quad x=0\\ 0 & \text{for} \quad x\to\infty\end{cases} \tag{2}$$



$dC(x)=nS\cdot G(x)dx$ $dC(x)=nSG(x)F(x)dx$

Fig 1 and 2: The general relationship between the source (which at the same time acts as the absorber) and the detector.

S is a known quantity. $G(x)$ can be known from either measurements or calculations. $C(h)$ is the quantity measured. Thus, the problem is to solve the integral equation (1) for n under the boundary condition (2).

First, we consider the trivial case where $C(h)$ can be obtained as an approximately continuous function of h. In this case, by differentiation of the equation (1) with h and using the condition (2), we can easily obtain n and $F(x)$.

$$n=\frac{1}{S}\frac{(\partial C/\partial h)_{h=o}}{G(o)} \tag{3-1}$$

$$F(x)=\frac{G(o)}{G(x)}\cdot\frac{(\partial C/\partial h)_{h=x}}{(\partial C/\partial h)_{h=o}} \tag{3-2}$$

Though this corresponds to the case of the strong intensity in which it is

not necessary to use a large quantity of source material, we can use these results for the aim of checking the validity of this method in the practical case.

In the practical cases, where the radioactive intensity may be very weak, we can use only several $C(h)$ at several h which have the meaningful differences, in taking into consideration the counting statistics and other experimental conditions. Thus, the integral equation (1) is replaced by the simultaneous equations on n and $F(x)$,

$$C(h_i)=nS\int_0^{h_i} G(x)\cdot F(x)dx \qquad (1')$$

$$i=1,\ 2,\ 3,\cdots$$

To solve these equations, some knowledge about the functional form of the function $F(x)$ is needed. Taking into account the fact that the attenuation factor of gamma-ray in the infinite and uniform medium can be expressed by the form $e^{-\mu x}(1+\delta x)$ for not so large distances (including the build-up factor), we can assume that $F(x)$ has a form of

$$F(x)=1+\sum_{j=1} p_j x^j$$

for not so large x. The validity of this assumption must be tested experimentally. As $G(x)$ is known quantity (by experiments or calculations), the equations (1′) can be reduced to a set of algebraic equations on n and p_j.

$$C(h_i)=nS\int_0^{h_i} G(x)dx+nS\sum_j p_j\int_0^{h_i} G(x)\cdot x^j dx \qquad (1'')$$

$$i=1,\ 2,\ 3,\cdots,\quad j=1,\ 2,\ 3,\cdots$$

So, we can get the value of n, by solving the equations (1″) or by using the least square method. As will be seen later, we can neglect all of the higher order terms except the first two, $1+p_1x$. Then, the least square method becomes the most preferable procedure.

Experimental Checks

(A) Appartus and Source

Source container was a cylinder of brass, the dimensions of which was 10.0 cm in inner diameter, 15.1 cm in depth and 0.12 cm in wall thickness.

The detector was a NaI scintillation crystal of $4''\phi\times 2''$ mounted upon the face of a photomultiplier (DuMont 6374). The output pulses were fed to the main amplifier and then to the RCL 256-channel pusle height analyzer.

As we had the object of determining the K^{40} content in the natural soil (Example II and III), we have used K^{42} (*12.52 h*) in the form of $K^{42}Cl$ as the radioactive material. Its gamma-ray energy is *1.53 Mev* and approximately equal to that of K^{40} (*1.46 Mev*). $K^{42}Cl$ was prepared in JRR-1 reactor of JAERI at Tokai.

(B) Example I

A small amount of the water solution of $K^{42}Cl$ was poured into the source container. The thickness was about *2.5 mm*, so the effects of the self-absorption could be perfectly negligible. The disintegration rate was absolutely determined by $\beta-\gamma$ coincidence method and found to be 1.4×10^4 *dps/cc*. By giving stepweis upward movements to the source container, $G(x)$ was determined experimentaly. Obtained $G(x)$ was very well realized by the linear combination of two exponential functions, which rendered the calculation of the definite integrals in (1″) very easy.

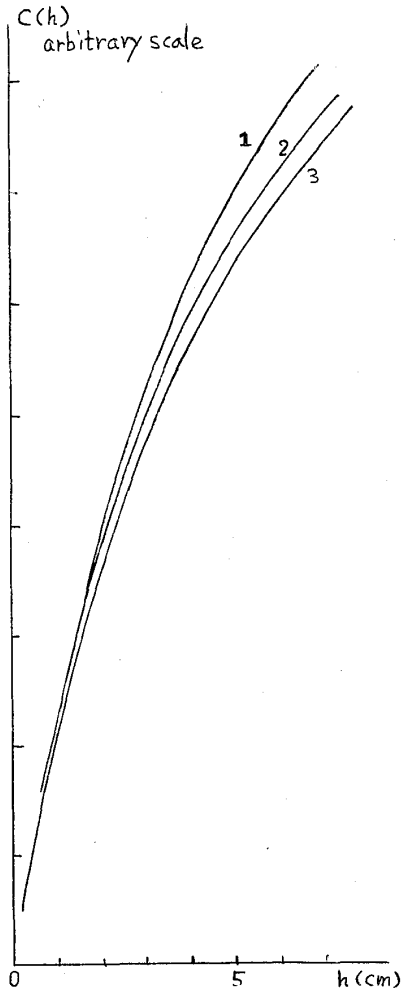


Fig 3: Accumulation curves for the gamma-ray in Example I.

Another *300 cc* water solution of $K^{42}Cl$ was divided into three equal parts. Each part was added to *900 cc* of H_2O (Sample 1), *270 g* $Pb(NO_3)_2$ solution in *900 cc* H_2O (Sample 2) and *455 g* $ZnCl_2$-$7H_2O$ solution in *900 cc* H_2O (Sample 3), respectively.

Sample 1 was poured into the initially empty source container stepweise and at each additions, counting of the photopeak area was recorded. The same procedures were carried on for other samples. Their accumulation curves are shown in Fig 3. By graphical differentiation, $F(x)$s were obtained and it was proved that the assumption $1+p_1x$ was a good ond.

n was determined in three different ways; 1) by the graphical differentiation based upon formula (3-1), 2) by solving the equations (1″) by selecting an arbitrary set of $C(h)$s at two heights, 3) by applying the least square method to an arbitrary set of $C(h)$s at three heights. Obtained results are tabulated in Table 1, which shows the usefulness of this

Table 1. The results of Example 1.

	in 10^4 *dps/cc*		
Methods	Sample 1	Sample 2	Sample 3
Numerical differentiation	*1.5*	*1.5*	*1.5*
Direct solving	*1.2*	*1.4*	*1.4*
Least square method	*1.3*	*1.3*	*1.3*

method.

(C) Example II

As the other test for the validity of this method, commercially available KCl powder was used as the source material. The radioactive material is isotope K^{40} in natural K. The apparatus and procedures were same as in Example I. Data are tabulated in Table 2. By applying the least square method, the K^{40} content in natural K was estimated to be *0.011%*, which was in good agreement to the true value, *0.012%*.

Table 2. K^{40} content in natural K.

Height (*cm*)	Weight (*g*)	Counting rate (*/s*)	*dps/g (KCl)*
4.2	*335*	*15.1±0.2*	*13.7*
7.2	*572*	*19.3±0.2*	
9.2	*735*	*20.1±0.2*	
11.7	*932*	*22.5±0.2*	

(D) Example III

About *1 Kg* of sand (sampled out at sea-shore in Zushi-City) was used as the source material. The radioactive material was isotope K^{40} in natural K contained in sand. The apparatus and procedures were same as in Example II. Data are tabulated in Table 3. By applying the least square method, the disintegration rate was estimated to be *0.25 dps/g (sand)*. In the conventional expression in which the weight ratio of K_2O against the sample is used, this value corresponds to *1.8%* which value can be taken as natural in the non-granite area.

Table 3. K content in natural sand.

Height (*cm*)	Weight (*g*)	Counting rate (*/s*)	*dps/g (sand)*
4.2	*527*	*0.503±0.02*	*0.25*
7.2	*879*	*0.663±0.02*	
11.7	*1378*	*0.842±0.02*	

Concluding Remarks

As is shown in Table 1 and 2, this method has a tendency of giving smaller values in comparison with the accurate values. The reasons are not yet clear. Even if we assume the form of $F(x)$ in $1+p_1x+p_2x^2$, no appreciable improvement has been obtained. It must be noted that, when the radioactive intensity per unit volume is very weak, the subtraction of the background

countings becomes very difficult. Because, in such cases, the source material itself plays a role of absorber for the background radiation and then may introduce some error in determining the true counting rate which might be comparable with the background counting rate.

The most advantageous point of this method is in the fact that arbitrariness exists only in assuming the form of $F(x)$ and, once it being done, there is no room into which arbitrariness enters. And also, the obtained accuracy of about ten percent may be a satisfactory one for most practically purposes.

We would like to express our sincere thanks to Prof. F. YAMAZAKI for his interest in this work.