SOME PROBLEMS CONCERNING THE CHOICE OF RADIOISOTOPES FOR TRANSMISSION TYPE MEASUREMENT METHODS

APPLICATION TO DENSITY MEASUREMENT OF STEAM — WATER MIXTURES

By

G. BEDE

Department for Power Stations, Technical University, Budapest

(Received October 18, 1969)

Presented by Prof. Dr. A. LÉVAI

Introduction

In radioactive measurements carried out on the principle of absorption the magnitude of the characteristics to be measured is generally deduced from the attenuation of the radiation occurring on the substance layer placed between the radioactive source and the detector. If e. g. a substance layer of a given thickness but of variable density is placed between the radioactive source and the radiation detector, the output signal of the detector and that of the gauge joined to the detector will vary according to the following rule (Fig. 1):

$$R = R_0 \exp\left[-\mu \varrho x\right] \tag{1}$$

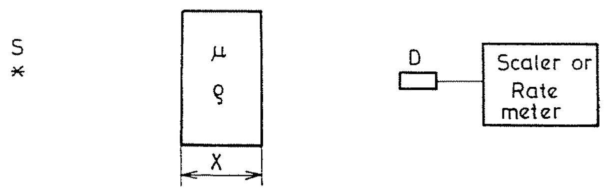

Fig. 1. D: Radiation detector; S: Radioactive source; μ: Mass absorption coefficient of the given medium (cm^2/g); ϱ: Density of the given medium (g/cm^3); X: Thickness of the sample (cm)

where R_0 is the output signal of the gauge if the geometry is unaltered but there is no substance to be measured (counts),

R is the output signal of the gauge for the substance to be measured (counts).

As seen, if ϱx varies $R(\varrho x)$ will vary exponentially.

According to the purpose of the measurement the method to be chosen should be the most suitable of the possible measurement methods based on the (1) relation in consideration of the following factors:

1) accuracy (sensitivity),
2) availability of the source,
3) costs of the system,
4) freedom from measurement interferences,
5) radiation safety considerations.

Considering that in industrial applications for example the density measurements are carried out on steamboilers under operation, only the γ and X-ray sources, respectively, can be taken into account of the possible sources as available because of their low attenuation and for radiation safety considerations. Therefore only these kinds of sources will be more intensively treated.

Factors affecting the choice of a γ (X-ray) source

The accuracy of the measurement method depends decisively on the radiation source applied in the measurement. Besides this decisive factor and the factors mentioned in 2), 3), however, the half-life of the source should also be taken into consideration.

In the followings these factors will be individually discussed.

1) *Accuracy (sensitivity) of the method*

In radioactive measurements the errors of the measurement results are fundamentally determined by two factors (the stable sources of errors as well as errors arisen from other sources being neglected), such as the random character of the radiation source's emission and the random deviations arisen from the uncertainty of the gauge. The effect of these factors can be expressed according to the well-known methods of the measurement technique, assumed that a variance in accordance of the Gauss-distribution is valid for both factors, as follows:

$$\sigma = (\sigma_s^2 + \sigma_i^2)^{1/2} \tag{2}$$

where σ is the standard deviation of the measured factor;

σ_s is the standard deviation of the statistic fluctuation of the source's emission;

σ_i is the standard deviation of the gauge's errors which vary statistically.

The gauge can be considered as a system, the output signal R of which being sort of a continuous function of the variable y to be measured:

$$R = f(y) \tag{3}$$

(In the present case e. g. $y = \mu \varrho x$ or, if the radiation source is given, $y = \varrho x$.) Consequently the inverse function of the relation (3), that is, the formula

$$y = F(R) \tag{4}$$

is the calibration function of the gauge.

Since in general the calibration function is not a straight line, neither the confidence intervals obtained around the function will be symmetrical to the function. It follows, that the value $[y \pm \sigma(y)]$ is generally not valid for the confidence intervals. If the value of $\sigma(R)$ is, however, small, R can be considered straight within the interval $(R - \sigma(R),\ R + \sigma(R))$ and therefore it is approximately true that the confidence intervals can be denoted by $[y \pm \sigma(y)]$. It is, however, also true within the given range that

$$\frac{\sigma(y)}{\sigma(R)} = \frac{\partial y}{\partial R} \tag{5}$$

and from this the $\sigma(y)$ being expressed:

$$\sigma(y) = \frac{\partial y}{\partial R} \cdot \sigma(R) \tag{5'}$$

or the same written in terms of the relative values

$$\frac{\sigma(y)}{y} = \frac{\partial y}{\partial R} \frac{\sigma(R)}{y}. \tag{6}$$

As shown by Eq. (2), the error of the measurement result can be disintegrated into two factors. The same carried out for $\sigma(y)$, determined in relation (6), shows that:

$$\frac{\sigma_s(y)}{y} = \frac{\partial y}{\partial R} \cdot \frac{\sigma_s(R)}{y} \tag{7}$$

and

$$\frac{\sigma_i(y)}{y} = \frac{\partial y}{\partial R} \cdot \frac{\sigma_i(R)}{y}. \tag{8}$$

As known the errors arisen from the statistical character of the radiation source's emission are different, depending on the type of the gauge, that is, the counter type gauge gives an error dissimilar to that of the ratemeter. In the former case [2]:

$$\sigma_s(R) = \sqrt{\frac{R}{t}} \tag{9}$$

(where t is the measurement time), and in the latter case:

$$\sigma_s(R) = \sqrt{\frac{R}{2\tau}} \tag{10}$$

(where τ is the time constant of the ratemeter). With help of relations (9) and (10) Eq. (7) can be rewritten for the case of a counter as

$$\frac{\sigma_s(y)}{y} = \frac{1}{y}\frac{\partial y}{\partial R}\sqrt{\frac{R}{t}} \tag{11}$$

and for the case of a ratemeter as

$$\frac{\sigma_s(y)}{y} = \frac{1}{y}\frac{\partial y}{\partial R}\sqrt{\frac{R}{2\tau}}\,. \tag{12}$$

Since the change ratio $\frac{\partial y}{\partial R}$ is the tangent slope of the calibration curve given by Eq. (4), in the knowledge of the calibration curve determined from the test data both $\sigma(y)$ and $\sigma_s(y)$ can be determined and then from Eqs (11) or (12) and (2) the effect of the gauge's error or better that of the statistical character of the source's emission on the error of the measured characteristics can be established. In routin measurements, if the choice of the radiation source had already been made, the value of σ_s can be reduced by the increase of the measurement time (or by the increase of the ratemeter's time constant). This effort is, however, frequently limited by the nature of the measurement.

The energy of the radiation source to be used for the measurement can also be optimized with help of Eqs (7) and (8). Supposed that $\sigma_i(y) \ll \sigma_s(y)$, the error of the measurement result is only characterized by $\sigma_s(y)$, a good approximation. Write

$$\sigma_y \simeq \frac{\partial y}{\partial R}\sqrt{\frac{R}{t}}\,. \tag{13}$$

(In the followings only the case of the counter gauge will be investigated. By the substitution of $t = 2\tau$ the result can, however, also be used for the ratemeter.) Transforming the relation (14) with help of relation (1) we get

$$\sigma(y) = -\frac{\exp\left[\mu\varrho x \cdot \frac{1}{2}\right]}{\mu\sqrt{tR_0}}\,. \tag{14}$$

The extreme of the function $\sigma(y) = f(\mu)$ given by Eq. (14) can be determined by the relation

$$\frac{\partial\sigma(y)}{\partial\mu} = O = \frac{1}{\sqrt{tR_0}} \exp\left[\frac{\mu\varrho x}{2}\right]\cdot\left[\frac{\varrho x}{2\mu} - \frac{1}{\mu^2}\right]. \tag{15}$$

As seen, the location of the extreme is

$$\mu = \frac{2}{\varrho x} \tag{16}$$

and the investigation of the second change ratio shows that the extreme is minimum.

Similar considerations, provided that $\sigma_s(y) \ll \sigma_i(y)$, the $\sigma_i(R)$ being no function of μ, lead to the conclusion that there exists again a minimum on the place of

$$\mu = \frac{1}{\varrho x}\,. \tag{17}$$

From the results of relations (16) and (17) conclusions can be drawn for the choice of the radiation source's energy.

In routin cases both of the sources of error (that is, the statistical character of the source's emission and the random error of the gauge, respectively) will occur. Consequently, the proper method of choice will be the use of a γ source (X-ray source) whose energy corresponds to the data of the given measurement problem (mass to be measured and geometry) and for which the condition

$$\frac{1}{\varrho x} < \mu < \frac{2}{\varrho x} \tag{18}$$

is valid. The condition (18) can only be satisfied graphically since, as known, the mass attenuation factor for γ-radiation depends both on the energy and on the atomic number of the given material. (It is to be noted that in case of β-radiation the mass attenuation factor is, with good approximation, only the function of the energy; with help of the above considerations the optimization of the radiation source's emission can be shown also in enclosed form [8].) The change of the mass attenuation factor as a function of the energy for the case of water is shown by Fig. 2 [6, 7].

The routin application of the above considerations is shown in connection with the density measurement of the water/vapour mixture flowing in the steam-generating tubes, one of the possible methods for the control of the circulation of steam-boilers of natural circulation. (The results of the density measurements, connected with data obtained by the measurement of other characteristics, provide an accurate control of the natural circulation [1].)

The density of the water/vapour mixture flowing through the steam-generating tubes of the steamboiler can be measured using the attenuation either of the radiation intensity in the mixture brought about by transmission or that of the energy.

Both of the phenomena can be expressed by Eq. (1). In our case the attenuation of the radiation intensity will be used as measurement method.

From relation (18), if the thickness of the layer to be transilluminated (that is, the internal diameter of the boiler's steam-generating tube) is 50 mm

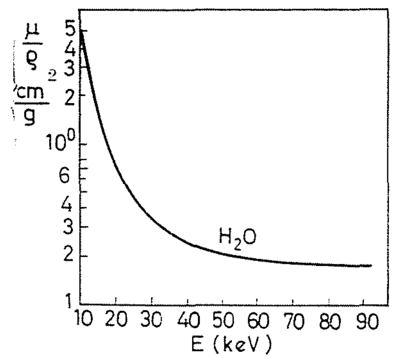

Fig. 2. Mass absorption coefficient for H_2O

and the range of density to be investigated is 0.5—1 g/cm³, the following conditions are given for the value of μ:

— at the lower limit of density	$0.4 < \mu < 0.8$	(19)′
— at the upper limit of density	$0.2 < \mu < 0.4$	(19)″
— commonly from both conditions	$0.2 < \mu < 0.8$	(19)‴

Fig. 2 shows that the condition (19) can be satisfied within the energy range of 18—60 keV by radiating γ sources. Several available γ sources are shown in Table 1 [3].

As shown in the Table, there exists a very small number of γ-radiating isotopes which would satisfy condition (19). Bremsstrahlung γ-sources will, however, easily satisfy condition (19). The energy spectra of some available Bremsstrahlung-sources are shown in Figs 3—5 [4, 5]. An estimation for the targets, which have not been indicated in the figures, is given in [5] according to which the peak of the Bremsstrahlung spectra is to be waited for the energy

$$E_b = A \cdot M^{0.2} \cdot Z^{0.7} \quad [\text{keV}] \tag{20}$$

Table 1

Data of several low-energy η -sources

Radiant substance	Energy of radiation keV	Half-life	Remark on the availability of the source
Ti^{44}	72 79	23 a	Radiation energy higher than the conditional zone
Fe^{60}	27	10^{5}a	From its disintegration arises Co^{60} not separable from Fe^{60} (β-disintegration)
As^{73}	13 54	76 d	
Kr^{81}	12	2.1 ^{5}a	Radiation energy lower than the conditional zone
Sn^{119m}	24 65	245 d	
Te^{127m}	59 89	105 d	The Te — 127 arisen from its disintegration emits at a higher energy (t $\frac{1}{2}$ = 9.5 h)
Sm^{145}	61	340 d	
Gd^{153}	70 97 103	236 d	Radiation energy higher than the conditional zone
W^{181}	92 136 152	145 d	Radiation energy higher than the conditional zone
Am^{241}	15 60	300 a	

where M denotes the thickness (g/cm^{2}) and Z the atomic number of the target, A is the empiric constant with a value of 8.6 for Sr^{90}—Y^{90} and 7.6 for P^{32}.

In our investigations carried out at the Department for Power Stations the Bremsstrahlung of Sr^{90}—Y^{90} β-radiation source brought about by targets of Al and Pb has been used. The γ-spectra, measured for a Pb target is shown in Fig. 6.

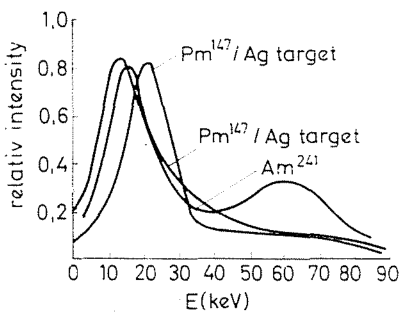

Fig. 3. Bremsstrahlung spectra [4]

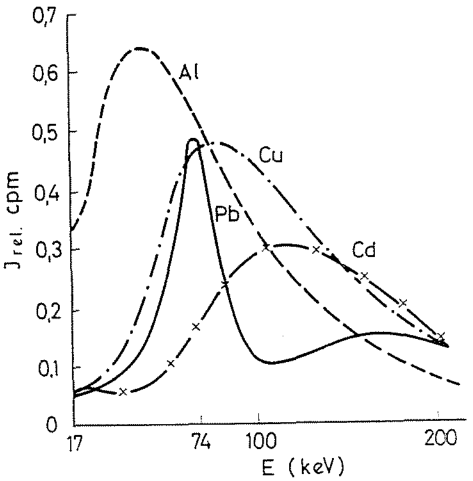

Fig. 4. Bremsstrahlung spectra for Sr^{90}—Y^{90} [5]

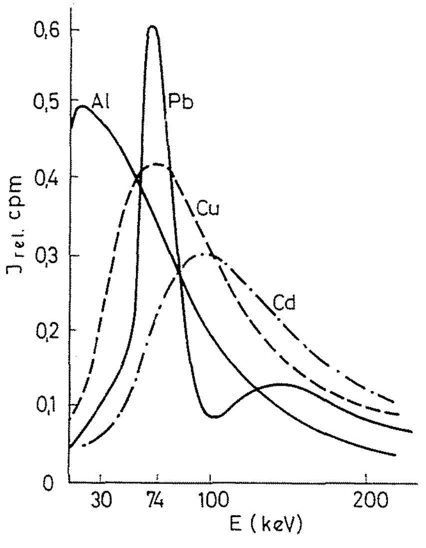

Fig. 5. Bremsstrahlung spectra for P^{32} [5]

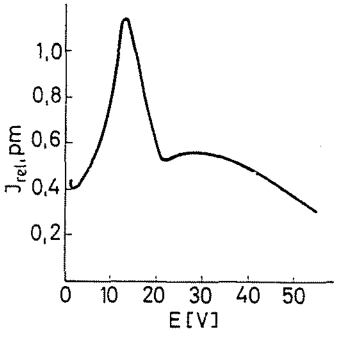

Fig. 6. Bremsstrahlung spectra for a $9Sr^{90}$—Y^{90} source (Pb target)

2) *Characteristics of the radiation source*

As mentioned, in addition to the requirements arisen in connection with the accuracy of the measurement, important factors of the proper choice are the accessibility, costs and half-life of the radiation source. These factors have been taken into consideration in Table 1, where the radiation sources with a half-life shorter than the duration of the measurement and those with a half-life are not comparable with the duration of the measurement are neglected. The number of the available isotopes continues to reduce if the accessibility and the price are considered, since Am-241 can only be imported from countries where nuclear fuel is being reprocessed and although As-73, Sm-147 and Sn-119m are produceable, there are no home experiences about their costs, similar to the other isotopes contained in Table 1.

In point of view of the above considerations the Bremsstrahlung sources are more favourable because of their long half-life ($>$ 30 years) and relatively low production costs. Bremsstrahlung sources, although not used in our theoretical investigations, can be successfully applied in in-situ measurements of very high temperature ($>$200 C° at the site of measurement), being imbedded in glass and as such resistant against the effects of high temperatures. As a disadvantage should be noted, however, that similarly to all Bremsstrahlung-sources their γ-radiation occurs as a fraction of the β-radiation. Several figures are shown in Table 2 [5].

Table 2

Radiation yield of Sr^{90}—Y^{90} Bremsstrahlung sources as the percentage of the β-radiation for different targets

Type of Bremsstrahlung source	Target (1 g/cm²)			
	Al, %	Cu, %	Cd, %	Pb, %
Transmission	2.9	2.8	2.3	2.2
Backscattering	1.2	1.8	2.8	5.9
Sandwich	3.9	4.2	3.9	4.2

These low values of the γ-radiation require that the activity of the source to be applied be several times 10 mCi, taking into consideration in addition to the yield also the attenuation of the beam during the transmission. This being, however, β-radiation, does not cause any essential problems as regards the radiation safety.

Summary

With radioactive measurements methods constraints can be established for the emission of the radiation source by means of separation of the errors arisen from the statistic character of the radiation source's emission and the random errors of the gauge.

For the measurement of the density of a water/vapour mixture the low-energy radioactive radiation source can reasonably be developed as a Bremsstrahlung source.

References

1. RÁDONYI, L.: Control of the boiler's circulation by γ transmission method. Thesis, inedited.
2. GARDNER, R. P.—ELY, R. J. Jr.: Radioisotope measurement applications in engineering. Reinhold, 1967.
3. LENGYEL—JÁSZ: Handbook for isotope-laboratory. Műszaki Kiadó, Budapest, 1967.
4. FILOSOFO, J. et al.: Design and characteristics of β-exited x-ray sources. Radioisotopes in the physical sciences and industry. vol. II, P. 3. IAEA, Proc. Series.
5. LEVEQUE, P. et al.: Studies and industrial applications of Bremsstrahlung for the β-rays of yttrium 90. Proc. ICPUAE **15**, 142 (1955).
6. BLATZ, H.: Radiation hygiene handbook. McGraw Hill, 1959.
7. LANDOLF—BÖRSTEIN: Atome und Ionen Bd., Teil 1, 6. Aufl. Berlin, 1950.
8. CAMERON, J. F.: Fluid density measurements in enclosed systems. Radioisotopes in scientific research. vol. I, p. 246. Pergamon Press, 1958.

Gábor BEDE, Budapest I., Fő u. 7, Hungary