Applied Radiation and Isotopes 109 (2016) 257–260

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Practical correction method for impurities on activity measurements using isotope calibrators

H. Ishizu *, T. Yamada

Japan Radioisotope Association, 28-45, Honkomagome 2, Bunkyo-ku, Tokyo 113-8941, Japan

HIGHLIGHTS

- Impurity correction methods were studied to determine activity concentration.
- Examples of activity measurements for ^{201}Tl and ^{64}Cu were shown.
- Two different shapes of responses were obtained using attenuation filter made of tin.
- The accuracy of the correction method with two different responses was discussed.

ARTICLE INFO

Article history:
Received 13 April 2015
Accepted 23 November 2015
Available online 26 November 2015

Keywords:
Ionization chamber
Impurity
Correction method
Activity measurement
Attenuation filter
^{201}Tl
^{202}Tl
^{64}Cu

ABSTRACT

Radioactive impurities might cause significant error in the activity determination of a target nuclide using ionization chambers. In the present study, an impurity correction technique for ^{201}Tl sources was performed by applying two different responses of an IG12A20 and IG11N20 ionization chamber. This technique can be extended to another method in which an attenuation filter made of tin was used to obtain different responses of an argon filled IG12A20. The results obtained with these techniques were very consistent with each other and with the reference value within their uncertainty after making the impurity correction. Examples of ^{64}Cu activity determination were also shown.

1. Introduction

Various kinds of radioisotopes such as ^{201}Tl and ^{64}Cu are widely used for many medical applications. Since the activity should be measured accurately to use these isotopes properly, various isotope calibrators employing a well-type ionization chamber (IC) are used in many hospitals.

However, radioactive impurities sometimes may be present in the source (e.g. Dryak et al. (1989)), causing significant error on the activity determination of the main nuclide on practical measurements using an isotope calibrator. For example, a ^{201}Tl source sometimes contains ^{202}Tl as a by-product. Since these nuclides have different half-lives, the activity ratio ^{202}Tl/^{201}Tl changes hour by hour. Therefore, in the inter-comparison of activity measurements for ^{201}Tl in hospitals, the indicated value of the isotope calibrator is dependent on its date of measurement (Baker and Woods, 2001).

Several types of impurity correction methods have been reported to solve such problems (Schrader, 1997). For example, correction method making use of different half-lives may be one of the useful techniques to eliminate the impurity components (Schrader and Walz, 1986). However, since a set of continual measurements over a few days shall be required with this method, a more practical method should be available for the routine measurement in hospitals. As other alternatives, two techniques using different shapes of response curves were studied for more practical approaches to correct the effects of radioactive impurities in routine radioactivity measurements with ICs. One uses two types of ICs each with different kinds of filling gases, *e.g.* argon and nitrogen gases, which give very different response shapes to each other especially in the lower energy region. The other employs an additional attenuation filter (inner liner) for argon filled IC. Use of an attenuation filter made of medium atomic number material, such as tin, is effective to reduce the inherent bump which appears in the lower energy region in the response curve of argon filled ICs.

In the present study, the three practical correction methods

* Corresponding author.
E-mail address: ishizu-hidetake@jrias.or.jp (H. Ishizu).

http://dx.doi.org/10.1016/j.apradiso.2015.11.083
0969-8043/

described above were tested for the correction of the influences of radioactive impurities contained in the ^{201}Tl sources, and the results obtained with these three methods were compared with each other. As another example, the impurity correction of ^{64}Cu was tried with the two ICs method with different responses.

2. Principles of three methods

2.1. Conventional correction method making use of different half lives

The difference of radioactive decays due to different half-lives is sometimes utilized for impurity corrections (Schrader and Walz, 1986). Generally, an indicated value of an isotope calibrator can be described for ^{201}Tl as the following equation.

$$A_{Tl-201} \times R_{Tl-201} \times \exp(\lambda_{Tl-201} \times t) + A_{Tl-202} \times R_{Tl-202} \times \exp(\lambda_{Tl-202} \times t) = S \quad (1)$$

where A_{Tl-201}, A_{Tl-202} are activities at reference time, R_{Tl-201}, R_{Tl-202} the responses to ICs, t the elapsed time after reference time, λ_{Tl-201}, λ_{Tl-202} the decay constants, and S the indicated value at each measurement time. Given the two indicated values S_1, S_2 at different measuring points t_1, t_2, Eq. (1) can be expressed as the following equation.

$$\begin{pmatrix} A_{Tl-201} \\ A_{Tl-202} \end{pmatrix} = \begin{pmatrix} R_{Tl-201} \times \exp(\lambda_{Tl-201} \times t_1) & R_{Tl-202} \times \exp(\lambda_{Tl-202} \times t_1) \\ R_{Tl-201} \times \exp(\lambda_{Tl-201} \times t_2) & R_{Tl-202} \times \exp(\lambda_{Tl-202} \times t_2) \end{pmatrix}^{-1} \begin{pmatrix} S_1 \\ S_2 \end{pmatrix} \quad (2)$$

If either A_{Tl-201}, A_{Tl-202} or R_{Tl-201}, R_{Tl-202} are known values, the other can be easily obtained by solving Eq. (2).

2.2. Correction method making use of two types of ICs with different response curves

Since the response curve of an IC depends on its design (*e.g.* filling gas, wall material, wall thickness etc.), two different shapes of responses are obtainable from two different types of ICs (*e.g.* IG12A20 and IG11N20). Since the responses at low energy region depend strongly on the kinds of filling gas, significant difference can be found between these two types of ICs (Lowenthal et al., 2001). Fig.1 shows the comparison of the response curves of argon-filled IC (IG11A20) and nitrogen-filled IC (IG11N20) which were obtained with the EGS-5 Monte-Carlo simulation code. In cases that the two measurements are made at nearly same time with two different types of ICs, the following equation can be obtained;

$$\begin{pmatrix} A_{Tl-201} \\ A_{Tl-202} \end{pmatrix} = \begin{pmatrix} R_{Tl-201} \times \exp(\lambda_{Tl-201} \times t) & R_{Tl-202} \times \exp(\lambda_{Tl-202} \times t) \\ R'_{Tl-201} \times \exp(\lambda_{Tl-201} \times t) & R'_{Tl-202} \times \exp(\lambda_{Tl-202} \times t) \end{pmatrix}^{-1} \begin{pmatrix} S_1 \\ S_2 \end{pmatrix}, \quad (3)$$

where R'_{Tl-201}, R'_{Tl-202} are responses to two different ICs. As represented in Eq. (3), each activity can be obtained quickly by solving Eq. (3) using known responses for ^{201}Tl and ^{202}Tl that can be obtained a priori from a reference solution containing ^{202}Tl as impurity.

2.3. Correction method using attenuation filter (inner liner)

The energy response of ICs can be changed by using an attenuation filter (inner liner) (Richard and O'brien, 1969; Schrader and Weiss, 1983). In particular, the use of an attenuation filter made of medium atomic number material such as tin is very effective to reduce the inherent bump appearing in around 100 keV region in the response of argon filled ICs. In this case, two different shapes of responses can be obtained using one isotope-calibrator, preferably with an argon filled IC. Consequently, Eq. (3) can be also applied even for the attenuation filter (inner liner) method.

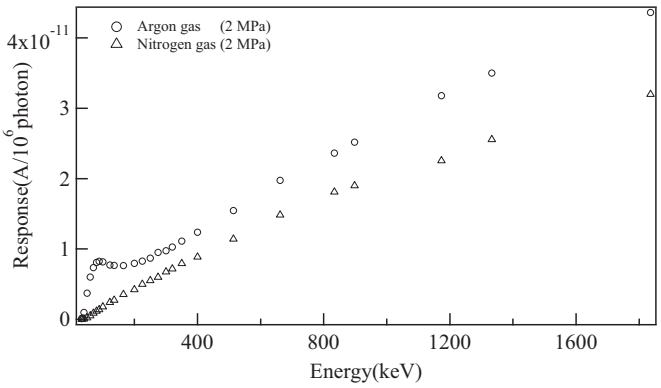

Fig. 1. Energy responses of IG11A20 and IG11N20 chambers as computed by EGS-5 Monte Carlo simulation code.

3. Experiments

3.1. Sample preparation of ^{201}Tl

For the experimental measurement, two kinds of ^{201}Tl source were prepared from the same ^{201}Tl solution; point sources for the activity determination using a calibrated Ge γ-ray spectrometer, and a solution source in a flame sealed ampoule (~5 ml) for IC measurements.

In order to prepare point sources, quantitative aliquots of the ^{201}Tl master solution (~20 MBq/g, carrier concentration: 0.1 mg/g TlCl in 0.1 N-HCl) were dropped on 0.8 mg/cm^2 Mylar films with a pycnometer, and then one drop of diluted colloidal silica solution (LUDOX SM-30, dilution factor; 8000) was added to reduce the self-absorption. After drying, each source was sandwiched with another Mylar film. This master solution, of approximately 5 ml, was filled in an ampoule made of Pyrex glass of 1 mm thickness and 18 mm of inner diameter. So far, this type of ampoule has been widely used in Japan. The mass of the solution was determined by differential weighing.

3.2. Measurements by a calibrated germanium spectrometer

The activity of each of the three point sources was measured using a calibrated germanium spectrometer, from which the ^{201}Tl activity concentration of the master solution was determined by dividing each activity by the deposited solution mass. The mean of the three results were adopted as the reference value. Small amount of ^{202}Tl and ^{200}Tl were detected as impurities in the present measurements, and their activity ratios ^{202}Tl/^{201}Tl and ^{200}Tl/^{201}Tl were determined to be 0.26% and 0.05%, respectively, at the reference time. No other impurities were detected. The activity ratios ^{202}Tl/^{201}Tl and ^{200}Tl/^{201}Tl should change in time according to their half-lives, and the ratios at any time can be calculated in a simple manner.

3.3. ^{201}Tl activity measurements using several ICs

The ^{201}Tl ampoule source was measured repeatedly in time for several types of ICs whose specifications are summarized in Table 1. Among the ICs used, the IG12A20 chamber with an inner liner made of tin of 1.25 mm thickness was also studied. Using the results, the apparent activity (indication) in terms of ^{201}Tl corrected at the reference time were plotted as a function of the activity ratios ^{202}Tl/^{201}Tl, which were determined by the manner