



The Standardisation of Samarium-153

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SUMMARY

Samarium-153 (^{153}Sm) is a beta-gamma emitting radionuclide. When it is labelled to a bone-seeking compound and administered to a patient with painful bone metastases it can provide palliative treatment of the disease. The purpose of this work was to produce an Australian primary standard of activity measurement for samarium-153. At the Australian Nuclear Science and Technology Organisation (ANSTO), the Australian primary standard method of activity is $4\pi\beta\text{-}\gamma$ coincidence counting. The activity determined would be used to calibrate secondary standards of activity measurement such as the Australian working standard method, a $4\pi\text{-}\gamma$ ionisation chamber. Two methods of activity determination were used. The first was efficiency extrapolation, where a drop of water was added to the source during counting. The efficiency steadily increased as the water dried and this was extrapolated to find the true disintegration rate. The second method used was tracer extrapolation. A tracer nuclide of known activity was added to the source. An efficiency extrapolation was performed as before and the tracer activity subtracted. Comparing the two methods showed the activity estimate of ^{153}Sm had an uncertainty of $\pm 3.5\%$. The activity and consequent calibration factors were compared with international standard methods for activity measurement.

1. INTRODUCTION

As yet there is no Australian primary standard method applied to the determination of activity for samarium-153. Current estimates of its activity could be in error by about 20%. The purpose of this work was to produce an Australian primary standard measurement of samarium-153. An activity estimate of samarium-153 using the Australian primary standard method is used as a calibration of secondary standard methods. For activity determination the absolute method of standardisation and the Australian primary standard method is the technique of $4\pi\beta\text{-}\gamma$ coincidence counting (1). The activity determined would be used to calibrate secondary standard methods such as the Australian working standard, the $4\pi\text{-}\gamma$ ionisation chamber.

2. METHODS

For $\beta\text{-}\gamma$ emitting radionuclides, a β particle is followed almost immediately by a γ -ray. The $4\pi\beta\text{-}\gamma$ coincidence counting system uses a detector for β radiation and a detector for γ radiation. A coincidence mixer registers a count when a pulse from the β detector arrives in coincidence with a pulse from the γ detector (2). The coincidence count rate relative to the β and γ count rates is a measure of the disintegration rate of the source. Figure 1 below illustrates the principle behind coincidence counting.

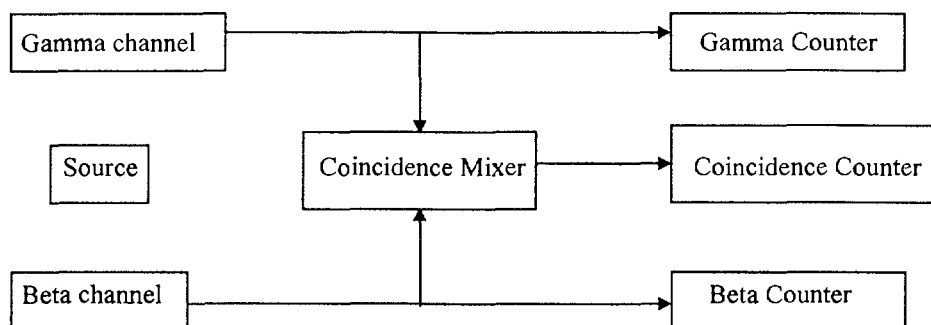


Figure 1. Simplified $\beta\text{-}\gamma$ coincidence setup (3)

The coincidence counting system at ANSTO uses a 4π steradian proportional counter for β particle detection and a NaI(Tl) crystal coupled to a photomultiplier tube for γ -ray detection (Figure 2). Ideally, the β detector would be

insensitive to γ radiation, and the γ detector would be insensitive to β radiation.

The disintegration rate (activity), N_0 , of a β - γ emitting radionuclide can be found by using the following equations (4):

$$N_\beta = N_0 \epsilon_\beta \quad (1)$$

$$N_\gamma = N_0 \epsilon_\gamma \quad (2)$$

$$N_c = N_0 \epsilon_\beta \epsilon_\gamma \quad (3)$$

where N_β is the observed β count rate
 N_γ is the observed γ count rate
 N_c is the observed coincidence count rate
 ϵ_β is the β detection efficiency (fraction of β particles observed)
 ϵ_γ is the γ detection efficiency (fraction of γ rays observed)

Rearranging equations 1-3:
$$N_0 = \frac{N_\beta N_\gamma}{N_c} \quad (4)$$

$$\epsilon_\beta = \frac{N_c}{N_\gamma} \quad (5)$$

Equation 4 shows that the absolute disintegration rate is independent of the efficiency of either detector.

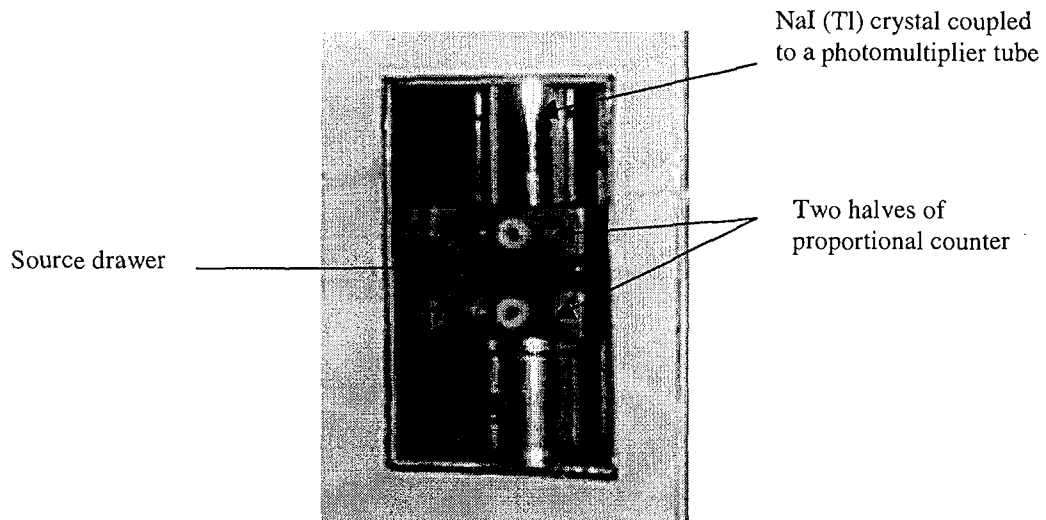


Figure 2. The detector system at ANSTO

2.1 Efficiency Extrapolation

The activity calculation will depend on the proportional counter efficiencies for conversion electrons, ϵ_{ce} , and γ rays, $\epsilon_{\beta\gamma}$ (4, 6). A correction formula for the proportional

counter response to conversion electrons and γ rays is given by (4, 6, and 7):

$$\frac{N_{\beta} N_{\gamma}}{N_c} = N_0 \left[1 + \frac{(1 - \epsilon_{\beta})}{\epsilon_{\beta}} \left(\frac{\alpha \epsilon_{ce} + \epsilon_{\beta\gamma}}{1 + \alpha} \right) \right] \quad (6)$$

where N_{β} and N_{γ} are the β and γ count rates corrected for background and dead times
 N_c is the coincidence count rate corrected for background and resolving times
 α is the internal conversion coefficient (the ratio of the number of conversion electrons to the number of γ rays) (3). Equation 6 can be simplified as follows (4, 5):

$$\frac{N_{\beta} N_{\gamma}}{N_c} = N_0 \left[1 + \frac{(1 - \epsilon_{\beta})}{\epsilon_{\beta}} \times K \right] \quad (7)$$

where
$$K = \frac{\alpha \epsilon_{ce} + \epsilon_{\beta\gamma}}{1 + \alpha} \quad (8)$$

The ^{153}Sm efficiency of the proportional counter to β radiation, ϵ_{β} , is at best about 90%.

If the efficiency was 100% ($\epsilon_{\beta} = 1$), then equation 7 would become:

$$\frac{N_{\beta} N_{\gamma}}{N_c} = N_0 \quad (9)$$

which is equation 4. It can then be seen that at $\epsilon_{\beta} = 1$, the value of K becomes irrelevant.

Equation 9 reduces further since $\epsilon_{\beta} = \frac{N_c}{N_{\gamma}}$,

then $N_{\beta} (\epsilon_{\beta} = 1) = N_0$.

$$N_{\beta} = N_0 f(\epsilon_{\beta}) \rightarrow N_0 \quad \text{as } \epsilon_{\beta} \rightarrow 1 \quad (10)$$

This is known as efficiency extrapolation (2, 5, 6, and 7). Efficiency dependent corrections include that due to self-absorption of the source (5, 8, and 9). The principle of efficiency extrapolation is such that any efficiency dependent correction is eliminated (5). One of the simplest ways to vary the β efficiency is to add a drop of water to the source just before

By varying the proportional counter efficiency, an efficiency function, $f(\epsilon_{\beta})$, may be extrapolated to $\epsilon_{\beta} = 1$, thus giving the true activity of the source. That is (2, 6):

counting (5). As the water evaporates, the efficiency (and count rate) will steadily increase. A plot of N_{β} versus $(1 - \epsilon_{\beta})/\epsilon_{\beta}$ can be extrapolated to $\epsilon_{\beta} = 1$ (that is $(1 - \epsilon_{\beta})/\epsilon_{\beta} = 0$) so that the β count rate is equal to the disintegration rate of the source ($N_{\beta} = N_0$).

2.2 Tracer Extrapolation

Tracer extrapolation involves the addition of a suitable β - γ emitting 'tracer' nuclide to the source (9, 10). The known activity of the tracer is subtracted from the total count rate to yield the activity of ^{153}Sm . The count rate is to be plotted as a function of the β -efficiency for the tracer nuclide (9). The efficiency tracing method was originally intended for the activity

determination of pure β -emitters (9). The γ channel SCA must then be gated on the γ energies of the tracer nuclide only (4).

In this work cobalt-60 (^{60}Co), a β - γ emitter, is used as the tracer nuclide. ^{60}Co emits γ rays of energies (1.17 & 1.33 MeV) much greater than that of ^{153}Sm . This has the advantage of

assuring that no γ ray from ^{153}Sm will contribute to the γ count rate if the γ window is set on the ^{60}Co peaks (4).

After the addition of ^{153}Sm onto a ^{60}Co source, a drop of water is added for an efficiency extrapolation. The β -efficiency, ϵ_β , is due to the ^{60}Co count rates only. The β count rate of the ^{60}Co - ^{153}Sm mixture is a function of both the β efficiency and the time of measurement, t . The ^{60}Co activity must be found as a function of the same ϵ_β and t . The β count rate due to

^{60}Co can be calculated by multiplying the β efficiency by the predetermined activity of ^{60}Co (3), using equation 1. The result is then subtracted from the $^{60}\text{Co}/^{153}\text{Sm}$ β count rate to reveal N_β of ^{153}Sm as a function of ϵ_β and t . This count rate may now be corrected for decay to a reference time. A plot of the decay corrected count rate versus an efficiency function can be extrapolated to give the true activity, N_0 .

3. RESULTS

Below is a comparison between efficiency extrapolation and tracer extrapolation.

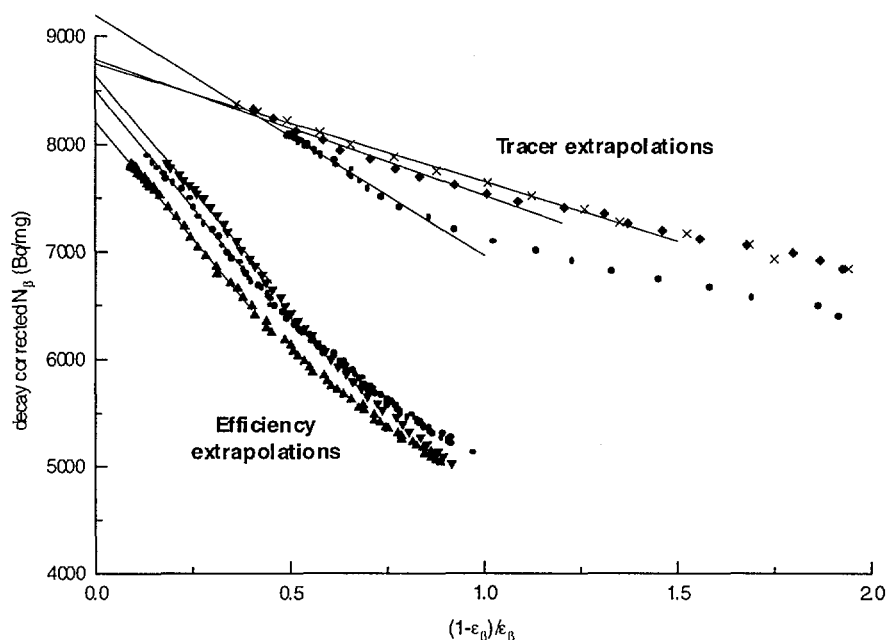


Figure 3. Comparing efficiency & tracer extrapolations for ^{153}Sm (12)

The mean and standard deviation of all the results yields an activity estimate of; $N_0 = 8.68 \pm 0.30$ MBq/g. The uncertainty here is $\pm 3.5\%$ at one standard deviation from the mean. A sample of ^{153}Sm was sent to the NPL (13), and another sent to NIST (14). The reported activities were compared with ion chamber

measurements calibrated using the Australian primary standard method of measurement. The results reported from NPL and NIST were found to be in disagreement by approximately 10%, with the Australian primary standard method falling between the two.

4. CONCLUSION

This work was based on the comparison between efficiency tracing and efficiency extrapolation. Both methods of activity

measurement are equally valid. Further work needs to be done to acquire an international standard of activity for samarium-153.

5. REFERENCES

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