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CALIBRATION AND OPERATION OF THE AAEC WORKING STANDARD OF MEASUREMENT FOR THE ACTIVITY OF RADIONUCLIDES
PART II : ION CHAMBER ANALYSIS

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ABSTRACT

As a secondary method for the determination of radioactivity, the ion chamber is both convenient and precise. The equations used in the calibration of the equipment and in the calculation of unknown activities are fully derived, and it is shown that the experimental procedure outlined is the optimum for both precision and convenience.

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ACCURACY; CALIBRATION; CALIBRATION STANDARDS; COBALT 60; ELECTRIC CURRENTS; HALF-LIFE; IONIZATION CHAMBERS; RADIOACTIVITY; RADIUM 226

CONTENTS

1. INTRODUCTION

The ion chamber equipment and procedure adopted at the Lucas Heights Research Laboratories has been described by Urquhart [1986]. The present report gives a complete derivation of the equations used for the calibration of the equipment, and for the calculation of the activities of samples of unknown activity. Further, it is shown that the procedure ensures the best precision and includes the most convenient form of calibration.

1.1 Determination of Radioactivity by the Ion Chamber

The absolute determination of radioactivity requires meticulous, time-consuming procedures which vary from one nuclide to another. A secondary method of determination is one in which the sample of unknown activity is measured in equipment which has been calibrated with sources whose activities were previously determined by an absolute method. The ion chamber provides a secondary method which is both precise and rapid. A general survey of the method is given in a report by the US National Council on Radiation Protection and Measurements [NCRP 1985].

1.2 Principle of the Method

The ionisation current produced by a radioactive source made from a particular radionuclide is proportional to the radioactivity (radioactive events per second) of that source. The specific ionisation current for the nuclide and ion chamber is defined as the ionisation current produced by a source of 1 MBq, and the unit of specific ionisation current is pA MBq^{-1}.

The ion chamber is calibrated by determining the specific ionisation current for each nuclide. These calibration factors are determined for a reference date which is the date of the original determination. If the ionisation current produced by a given source of unknown radioactivity is then measured, the radioactivity can be calculated. This method has to be modified to take into account changes which may take place in the equipment between the time of calibration and the time when the sample source is measured. The modification involves the use of radium-226 reference sources.

If a radium reference source was not in secular equilibrium at the time of the original set of calibrations, its calibration factor (or ionisation current) can be calculated for that date from subsequent measurements. This has an additional benefit, namely that when later samples are measured, there is a reduction in the uncertainty arising from the uncertainty in the half-life of radium.

The ionisation current is determined by an electrometer. The time interval in which the ionisation current raises the potential difference across the integrating capacitor from one set level to another is measured. Let C denote the capacitance of the capacitor which is being charged by the ionisation current I. Let ΔV denote the increase in potential difference across the capacitor in the time interval Δt. Then

$$I = C \frac{\Delta V}{\Delta t} .$$

2. DERIVATION OF THE ION CHAMBER EQUATION

2.1 Ion Chamber with Constant Sensitivity

Let $A_{\sigma o}$ denote the radioactivity of the source used to calibrate the ion chamber for a particular nuclide, and suppose that $A_{\sigma o}$ was measured by an absolute method (*e.g.* coincidence counting) at time t_o. Note that where two subscripts are used, the first refers to the source and the second to the time of measurement; thus σ denotes a standard source, and O (zero) a measurement at time t_o.

Subsequent to time t_o, suppose that the ion chamber is calibrated with the standard source at time t_c. At this time, the radioactivity ($A_{\sigma c}$) of the standard source will be given by

$$A_{\sigma c} = A_{\sigma o} \ exp \left[- \frac{ln 2}{T_n} (t_c - t_o) \right] ,$$

where T_n is the half-life of the particular nuclide. (In the symbol T_n, T denotes half-life, and the subscript refers to the nuclide.)

Let the specific ionisation current of the nuclide n at time t_c be denoted by i_{nc}. Let I_{bc} be the background ionisation current (*i.e.* with no source in the chamber) at time t_c, and let $I_{\sigma c}$ be the ionisation current produced by the standard source and background radiation at time t_c. Then, by the definition of i_{nc},

$$i_{nc} = \frac{I_{\sigma c} - I_{bc}}{A_{\sigma c}} . \qquad 2.1$$

Suppose that at a later time t_m a sample of the same nuclide, of unknown radioactivity A_{sm}, produces (with background radiation) an ionisation current I_{sm}, and that the background current is I_{bm}. The specific ionisation current is denoted by i_{nm}, with

$$i_{nm} = \frac{I_{sm} - I_{bm}}{A_{sm}} . \qquad 2.2$$

If the specific ionisation current has not changed between times t_c and t_m, *i.e.* $i_{nm} = i_{nc}$, then A_{sm} can be calculated from equation 2.2. However, the sensitivity of the chamber may have changed, so that i_{nm} does not equal i_{nc}. This difficulty is overcome by the use of a radium reference source.

2.2 Ion Chamber with Sensitivity Subject to Drift

Consider now the effect of change of sensitivity of the ion chamber. Let i_{rc} and i_{rm} denote the specific ionisation currents for the radium reference source at times t_c and t_m, respectively. It is assumed that as the chamber sensitivity changes, the specific ionisation currents of the sample nuclide and the radium reference source change in the same proportion, *i.e.*

$$\frac{i_{nm}}{i_{nc}} = \frac{i_{rm}}{i_{rc}}$$

$$= \frac{(I_{rm} - I_{bm}) / A_{rm}}{(I_{rc} - I_{bc}) / A_{rc}} ,$$

where I_{rm} and I_{rc} are the currents produced by the radium reference source and background radiation at times t_m and t_c, respectively, and A_{rm} and A_{rc} are the activities of the radium reference source at those times.

$$A_{rm} = A_{rc} \, exp \, [- \frac{ln 2}{T_r} (t_m - t_c)] ,$$

where T_r is the half-life of the radium reference source. Therefore

$$i_{nm} = \frac{(I_{rm} - I_{bm}) \, i_{nc}}{(I_{rc} - I_{bc}) \, exp \, [- \frac{ln 2}{T_r} (t_m - t_c)]} \qquad 2.3$$

Rearranging equation 2.2,

$$A_{sm} = \frac{I_{sm} - I_{bm}}{i_{nm}} ,$$

and substituting from equation 2.3 we get

$$A_{sm} = \frac{(I_{sm} - I_{bm}) \, (I_{rc} - I_{bc}) \, exp \, [- \frac{ln 2}{T_r} (t_m - t_c)]}{(I_{rm} - I_{bm}) \, i_{nc}} .$$

Substituting from equation 3.1,

$$A_{sm} = \frac{(I_{sm} - I_{bm})(I_{rc} - I_{bc}) \, A_{\sigma c} \, exp \, [- ln \frac{2}{T_r} (t_m - t_c)]}{(I_{rm} - I_{bm}) \, (I_{\sigma c} - I_{bc})} . \qquad 2.4$$

Suppose that the same capacitor and nominal potential difference have been used during the above measurements. We assume that although C and ΔV may change with time, the change is negligible during the calibration at time t_c and during the measurement of the sample of the nuclide carried out at time t_m. Let the values of C and ΔV at time t_c be C_c and ΔV_c; let the values at time t_m be C_m and ΔV_m:

$$I_{sm} = C_m \frac{\Delta V_m}{\Delta t_{sm}} \qquad I_{bm} = C_m \frac{\Delta V_m}{\Delta t_{bm}} \qquad I_{rm} = Cm \frac{\Delta V_m}{\Delta t_{rm}}$$

$$I_{\sigma c} = C_c \frac{\Delta V_c}{\Delta t_{\sigma c}} \qquad I_{bc} = C_c \frac{\Delta V_c}{\Delta t_{bc}} \qquad I_{rc} = C_c \frac{\Delta V_c}{\Delta t_{rc}}$$

Substituting in equation 2.4, and cancelling out C_m ΔV_m and C_c ΔV_c, we obtain

$$A_{sm} = \frac{(1/\Delta t_{sm} - 1/\Delta t_{bm})}{(1/\Delta t_{rm} - 1/\Delta t_{bm})} \times (1/\Delta t_{rc} - 1/\Delta t_{bc}) \times \frac{A_{\sigma c}}{(1/\Delta t_{\sigma c} - 1/\Delta t_{bc})} \times \exp\left[- \frac{\ln 2}{T_r} (t_m - t_c) \right] \tag{2.5}$$

For calculating A_{sm} we can tabulate the factor $(1/\Delta t_{rc} - 1/\Delta t_{bc})$ for all the reference sources measured at time t_c, and the factor

$$\frac{1/\Delta t_{\sigma c} - 1/\Delta t_{bc}}{A_{\sigma c}}$$

for the standard sources of various nuclides measured at time t_c.

It can be seen from the derivation of equation 2.5 that although C and ΔV must not vary during the calibration or the measurement of the sample, changes *between* times t_c and t_m do not matter because the values of C and ΔV cancel out in the equation; thus it is not necessary to know C and ΔV exactly.

If C_c ΔV_c is not cancelled out in equation 2.5, then

$$A_{sm} = \frac{1/\Delta t_{sm} - 1/\Delta t_{bm}}{1/\Delta t_{rm} - 1/\Delta t_{bm}} \times \frac{I'_{rc}}{i_{nc}} \times \exp\left[- \frac{\ln 2}{T_r} (t_m - t_c) \right] \tag{2.6}$$

where

$$I'_{rc} = I_{rc} - I_{bc} = C_c \Delta V_c \; (1/\Delta t_{rc} - 1/\Delta t_{bc}) \tag{2.7}$$

and

$$i_{nc} = \frac{I_{\sigma c} - I_{bc}}{A_{\sigma c}} = \frac{C_c \Delta V_c \; (1/\Delta t_{\sigma c} - 1/\Delta t_{bc})}{A_{\sigma c}} \tag{2.8}$$

For calculating A_{sm} we can tabulate I'_{rc} and i_{nc} instead of the factors mentioned above. When calculating I'_{rc} and i_{nc}, only approximate values of C_c and ΔV_c need be used because $C_c \Delta V_c$ cancels out when A_{sm} is calculated from equation 2.6.

2.3 Determination of Background

The interval Δt_{bc} (due to background) is too long for a direct determination, and is therefore determined as follows. A time interval $(\Delta t'_{bc})$ is measured when no source is present at time t_c and with a small capacitor (C'_c) and a small potential difference $(\Delta V'_c)$ selected:

$$\frac{C_c \Delta V_c}{\Delta t_{bc}} = \frac{C'_c \Delta V'_c}{\Delta t'_{bc}} \; (= I_{bc}) \; . \tag{2.9}$$

Δt_{bc} can then be calculated from the above equation. Because $1/\Delta t_{bc}$ (in equations 2.5, 2.7 and 2.8) is small, no appreciable error will occur in the calculation of A_{sm} if C_c, ΔV_c, C'_c and $\Delta V'_c$ are not known accurately. Δt_{bm} is obtained in the same way.

2.4 Choice of Working Equation

In calculating A_{sm}, which is the activity of a given sample, the most straightforward method is to use equation 2.5 together with tabulated values of the calibration factors:

$$1/\Delta t_{rc} - 1/\Delta t_{bc} \; ; \; \frac{1/\Delta t_{\sigma c} - 1/\Delta t_{bc}}{A_{\sigma c}} \; .$$

However, the practice in our laboratory is to tabulate the currents I'_{rc} and i_{nc}; the working equation is therefore equation 2.6.

2.5 Minimisation of Effect of Background Uncertainty

Let us consider the effect on accuracy of a change in background between the time when the interval Δt_{bm} is measured and the time shortly afterwards when Δt_{sm} and Δt_{rm} are measured. We shall assume that no change in background takes place during the latter two measurements. Thus equation 2.5 still applies but Δt_{bm} is incorrect. We shall now consider A_{sm}, the activity of a given sample, as a function of Δt_{bm}, with the other variables made constant for the time being. The effect on A_{sm} of a small change in Δt_{bm} will be least when the derivative of A_{sm} with respect to Δt_{bm} is zero, *i.e.* when

$$\frac{\partial A_{sm}}{\partial \Delta t_{bm}} = 0 \ . \qquad 2.10$$

Differentiating equation 2.5 and substituting into equation 2.10, we get

$$\frac{(1/\Delta t_{rm} - 1/\Delta t_{sm})}{\Delta t_{bm}^{2}\ (1/\Delta t_{rm} - 1/\Delta t_{bm})^{2}} \times (1/\Delta t_{rc} - 1/\Delta t_{bc}) \times \qquad 2.11$$

$$\frac{A_{\sigma c}}{(1/\Delta t_{\sigma c} - 1/\Delta t_{bc})} \times$$

$$exp\ [-\frac{ln 2}{T_r}\ (t_m - t_c)\ ] = 0$$

Equation 2.11 is satisfied if $\Delta t_{rm} = \Delta t_{sm}$. Thus, by preparing a sample which will produce the same ionisation current as the radium reference source, we minimise the effect on A_{sm} of any uncertainty in Δt_{bm}. The Δt_{bm} uncertainty may, as was noted above, be due to an actual change in background, or it may be due to uncertainties in capacitances and potential differences, as mentioned in section 2.3. Similarly, the effect of uncertainty in Δt_{bc} is minimised if $\Delta t_{rc} = \Delta t_{\sigma c}$.

3. RECALIBRATION OF THE ION CHAMBER

3.1 Recalibration with a Standardised Source at Time t_m

Suppose that the source with radioactivity A_{sm} has been prepared from a solution which has just been standardised by an absolute method. This source can then be used to recalibrate the ion chamber at time t_m. If, at a later time t_p, a source of unknown radioactivity (A_{zp}) is measured, then A_{zp} is calculated by substituting the new values in equation 2.5 (writing z for s, p for m, s for σ, and m for c):

$$A_{zp} = \frac{(1/\Delta t_{zp} - 1/\Delta t_{bp})}{(1/\Delta t_{rp} - 1/\Delta t_{bp})} \times (1/\Delta t_{rm} - 1/\Delta t_{bm}) \times \qquad 3.1$$

$$\frac{A_{sm}}{1/\Delta t_{sm} - 1/\Delta t_{bm}} \times exp\ [-\frac{ln 2}{T_r}\ (t_p - t_m)] \quad .$$

Because the time between calibration and sample measurement has been reduced from (t_p - t_c) to (t_p - t_m), the uncertainty in A_{zp} due to any uncertainty in T_r is also reduced.

3.2 Recalibration Referred to the Reference Time t_c

The results obtained at time t_m with the new standard source of radioactivity A_{sm} can also be used to calculate a nuclide calibration factor for time t_c. If this is done, the calibration factors of the reference sources need to be tabulated for t_c only, thereby minimising tabulation. It is shown in this section that accuracy is not affected by calculating the nuclide calibration factor in this way. We can now refer to t_c (the date of the original calibrations) as the reference time.

Instead of the calibration factor

$$\frac{1/\Delta t_{\sigma c} - 1/\Delta t_{bc}}{A_{\sigma c}}$$

being calculated from a measurement on a standard source at time t_c, it is obtained by rearranging equation 2.5 :

$$\frac{1/\Delta t_{\sigma c} - 1/\Delta t_{bc}}{A_{\sigma c}} = \frac{(1/\Delta t_{sm} - 1/\Delta t_{bm})}{(1/\Delta t_{rm} - 1/\Delta t_{bm})} \times \qquad 3.2$$

$$\frac{(1/\Delta t_{rc} - 1/\Delta t_{bc})}{A_{sm}} \times exp\ [-\frac{ln 2}{T_r}\ (t_m - t_c)] \ ,$$

where A_{sm} is known from an absolute determination, and the time intervals on the right-hand side of the equation have been measured at times t_c and t_m.

Suppose for the time being that T_r is known exactly. The radioactivity A_{zp}, which is to be determined at time t_p, is calculated by first substituting into equation 2.5 the time intervals for the sample and the reference source obtained at times t_p and t_c :

$$A_{zp} = \frac{(1/\Delta t_{zp} - 1/\Delta t_{bp})}{(1/\Delta t_{rp} - 1/\Delta t_{bp})} \times (1/\Delta t_{rc} - 1/\Delta t_{bc}) \times \frac{A_{\sigma c}}{(1/\Delta t_{\sigma c} - 1/\Delta t_{bc})} \times exp\,[-\frac{ln\,2}{T_r} (t_p - t_c)] \qquad 3.3$$

Suppose again that T_r is known exactly. Substituting in the above equation the expression for

$$\frac{1/\Delta t_{\sigma c} - 1/\Delta t_{bc}}{A_{\sigma c}}$$

given by equation 3.2, we obtain

$$A_{zp} = \frac{(1/\Delta t_{zp} - 1/\Delta t_{bp})}{(1/\Delta t_{rp} - 1/\Delta t_{bp})} \times (1/\Delta t_{rc} - 1/\Delta t_{bc}) \times \frac{(1/\Delta t_{rm} - 1/\Delta t_{bm})}{(1/\Delta t_{sm} - 1/\Delta t_{bm})} \times \frac{A_{sm}}{(1/\Delta t_{rc} - 1/\Delta t_{bc})} \times \frac{exp\,[- \frac{ln\,2}{T_r} (t_p - t_c)]}{exp\,[- \frac{ln\,2}{T_r} (t_m - t_c)]} \qquad 3.4$$

Simplifying, we obtain an equation which is identical with equation 3.1.

Suppose that in equations 3.2 and 3.3 T_r is replaced by T_{Ra}, which is a published value of the half-life of radium. The error in T_{Ra} will give rise to an error in

$$\frac{1/\Delta t_{\sigma c} - 1/\Delta t_{bc}}{A_{\sigma c}}$$

calculated by equation 3.2.

However, since substituting the above expression in equation 3.3 gives us equation 3.1 with T_r replaced by T_{Ra}, the error in A_{zp} is not increased by having used the above expression as the calibration factor for the nuclide instead of

$$\frac{1/\Delta t_{sm} - 1/\Delta t_{bm}}{A_{sm}}.$$

For the sake of simplicity, t_c is always adopted as a 'reference' time (as shown above) when the ion chamber is recalibrated, or calibrated for a new nuclide. We can either tabulate

$$\frac{1/\Delta t_{\sigma c} - 1/\Delta t_{bc}}{A_{\sigma c}}$$

as the calibration factor for the nuclide, or tabulate i_{nc}, which is the specific ionisation current of the nuclide.

As for the radium reference sources, it is necessary, for the calculation of the activity of subsequent samples, to tabulate only the calibration factors

$$(1/\Delta t_{rc} - 1/\Delta t_{bc})$$

or the ionisation currents (I'_{rc}) for one particular time only, namely t_c.

4. SECULAR EQUILIBRIUM AND HALF-LIFE OF THE REFERENCE SOURCE

4.1 Recalibration after Reference Source Attains Secular Equilibrium

The derivation of equations 2.5 and 2.6 depends on the validity of the equation

$$A_{rm} = A_{rc} \; exp \left[- \frac{ln2}{T_r} \; (t_m - t_c) \right] \qquad 4.1$$

in the period between t_c and t_m. However, this equation is valid only if the reference source, consisting of radium and its decay products in a sealed platinum capsule, has attained secular equilibrium before time t_c.

Thus Δt_{rc} should be measured with the reference source in secular equilibrium. Suppose that secular equilibrium is not well established until some time between t_c and t_m. The activity A_{rc} at time t_c will not then be given by rearrangement of equation 4.1, namely

$$A_{rc} = A_{rm} \; exp \left[\frac{ln2}{T_r} (t_m - t_c) \right] \qquad 4.2$$

and the time interval actually measured with the reference source in the ion chamber at t_c will be unsuitable for calculating

$$(1/\Delta t_{rc} - 1/\Delta t_{bc})$$

and I'_{rc}.

However

$$(1/\Delta t_{rc} - 1/\Delta t_{bc})$$

can be determined at time t_m in the manner described below, and used later in the measurement of samples of unknown radioactivity. Rearranging equation 2.5, we get

$$1/\Delta t_{rc} - 1/\Delta t_{bc} = A_{sm} \times \frac{(1/\Delta t_{rm} - 1/\Delta t_{bm})}{(1/\Delta t_{sm} - 1/\Delta t_{bm})} \times \frac{(1/\Delta t_{\sigma c} - 1/\Delta t_{bc})}{A_{\sigma c}} \times \frac{1}{exp \left[- \frac{ln 2}{T_r} \; (t_m - t_c) \right]} \qquad 4.3$$

Suppose that for the time being, T_r is known exactly. The radioactivity A_{sm} is determined by an absolute method just before t_m; $A_{\sigma c}$ will have been similarly determined already. The time intervals on the right hand side of equation 4.3 having been measured,

$$(1/\Delta t_{rc} - 1/\Delta t_{bc})$$

can be calculated from equation 4.3; I'_{rc} can then be calculated from equation 2.7.

Thus, even though calibration measurements may have been carried out on standardised sources of various nuclides before one or more of the radium reference sources had attained secular equilibrium, that work is not wasted, because later, when secular equilibrium has been attained, the necessary calibration factor or I'_{rc} can be obtained by making further measurements on the reference sources and 'calculating back'.

4.2 Effect of Uncertainty of Half-life on Recalibration

Let us see what effect the above method of determining

$$(1/\Delta t_{rc} - 1/\Delta t_{bc})$$

and I'_{rc} has on the accuracy of subsequent ion chamber results when the uncertainty in the half-life of radium is taken into account. Suppose we consider the general case of a measurement carried out on a source of another radionuclide. Suppose that at time t_c, its calibration source, with radioactivity $A_{\sigma' c}$, gave rise to a time interval of $\Delta t_{\sigma' c}$, and suppose that at time t_p (later than t_m) a sample of the nuclide of unknown radioactivity $A_{z' p}$ gave rise to a time interval of $\Delta t_{z' p}$.

$A_{z'p}$ is calculated in the following manner. We substitute into equation 2.5 the symbols in the last sentence and the time intervals Δt_{bp} and Δt_{rp}:

$$A_{z'p} = \frac{(1/\Delta t_{z'p} - 1/\Delta t_{bp})}{(1/\Delta t_{rp} - 1/\Delta t_{bp})} \times (1/\Delta t_{rc} - 1/\Delta t_{bc}) \times \frac{A_{\sigma' c}}{(1/\Delta t_{\sigma' c} - 1/\Delta t_{bc})} \times exp\,[-\frac{ln2}{T_r}\;(t_p - t_c)] \qquad 4.4$$

In the above equation, we now substitute for

$(1/\Delta t_{rc} - 1/\Delta t_{bc})$

the expression given in equation 4.3, and then combine the two *'exp'* factors:

$$A_{z'p} = \frac{(1/\Delta t_{z'p} - 1/\Delta t_{bp})}{(1/\Delta t_{rp} - 1/\Delta t_{bp})} \times A_{sm} \times \frac{(1/\Delta t_{m} - 1/\Delta t_{bm})}{(1/\Delta t_{sm} - 1/\Delta t_{bm})} \times \frac{(1/\Delta t_{\sigma c} - 1/\Delta t_{bc})}{A_{\sigma c}} \times \frac{A_{\sigma' c}}{(1/\Delta t_{\sigma' c} - 1/\Delta t_{bc})} \times exp\,[-\frac{ln2}{T_r}\;(t_p - t_m)\;] \qquad 4.5$$

If we substitute T_{Ra} for T_r in the decay factor of the above equation, we see that the accuracy of the determination of $A_{z'p}$ has been improved because the decay factor contains $(t_p - t_m)$ instead of $(t_p - t_c)$ which is larger. Any change in the sensitivity of the chamber between times t_c and t_m has been determined not by the radium reference source, but by the measurement of the time intervals of the two sources with radioactivities $A_{\sigma c}$ and A_{sm} that have been determined by an absolute method.

Thus the redetermination of

$(1/\Delta t_{rc} - 1/\Delta t_{bc})$,

apart from considerations of secular equilibrium, reduces the uncertainty in ion chamber measurements arising from the uncertainty in value of the half-life of radium.

5. SUMMARY OF CALIBRATION PROCEDURES

5.1 Calibration for a New Radionuclide

To obtain the calibration factor referred to time t_c for a new radionuclide, time interval measurements are made on an ampoule containing a solution of the nuclide which has recently been standardised by an absolute method. The factor is calculated from equation 3.2.

5.2 Recalibration of a Reference Source

Recalibration of a reference source is carried out by measuring time intervals for the reference source and for an ampoule containing a recently standardised solution of cobalt-60. Using these results and those obtained on an ampoule of standardised cobalt-60 solution at time t_c, the calibration factor for the reference source at time t_c is calculated from equation 4.3. The same procedure is used for the calibration of a new reference source.

6. REFERENCES

NCRP [1985] - A handbook of radioactivity measurements procedures. National Council on Radiation Protection and Measurements. NCRP Report No.58

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