Standardisation of pure β and β–γ emitting radionuclides in various physical forms

Li Mo

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Li Mo
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Institute of Medical Physics
The University of Sydney

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This thesis is submitted to the University of Sydney in fulfilment of the requirement for the Degree of Doctor of Philosophy.

The work presented in this thesis, to the best of my knowledge and belief, original except as acknowledged in the text. I hereby declare that I have not submitted this material, either in full or in part, for a degree at this or any other institution.

Signature:  

Date:
Abstract

The motivation of this study is user demand, from radiopharmaceutical industries and nuclear engineering, for accurate activity standards of radionuclides with different types of decay scheme and in various physical forms.

Firstly, this thesis describes the recently developed techniques in liquid scintillation counting, TDCR and CIEMAT/NIST methods, for the activity standardisation of pure β-emitters. Both theoretical and experimental studies on the influence of the single photoelectron peak are carried out on pure β-emitters $^3$H, $^{63}$Ni, $^{14}$C and $^{90}$Sr/$^{90}$Y. A modified formula for calculation of detection efficiency is developed in consideration of the rejected fraction of the single photoelectron peak.

TDCR and CIEMAT/NIST methods are then applied to standardise an important therapeutic radiopharmaceutical, $^{90}$Y microspheres, which is used in the treatment of liver cancer. The technique developed in this study to deal with this solid source is the highly quantitative chemical digestion of the microspheres.

$4\pi\beta-\gamma$ coincidence counting technique is the most powerful technique for the absolute activity measurements of β-γ emitting nuclides. However it is used to measure ultra-thin sources. This thesis describes the study carried out in extending this technique to thick solid sources, Au wires, Al-Au wires and Au foils. The demand for standardisation of $^{198}$Au in the form of Au wires, Al-Au wires and Au foils is from the hot commissioning of the OPAL (Open Pool Australian Light-Water) nuclear reactor at the Australian Nuclear Science and Technology Organisation (ANSTO). A Monte Carlo simulation was employed to simulate the complicated absorption and attenuation processes of electrons and gamma photon interactions with the surrounding materials. Probabilities of escape beta particles, internal conversion electrons and photon-interaction generated photoelectrons and Compton electrons calculated using the Monte Carlo simulation were used to determine the correction term of the coincidence equation. The successful use of the activity standards of $^{198}$Au in the form of Au wires and Al-Au wires in the neutron flux measurements for the hot commissioning process of the ANSTO OPAL reactor has proved the accuracy of the standards developed in this work.

The calibration of the dose calibrator used by the Australian National Medical Cyclotron (NMC) Positron Emission Tomography (PET) Quality Control (QC) Section for the $^{18}$F carried out in this work ensures the activity measurement of this radiopharmaceutical traceable to the Australian primary standard. The establishment of Australian secondary dosimetry standard for air kerma rate measurement of $^{125}$I brachytherapy seeds ensures the dose rate measurement is traceable to an international standard.
Finally, in order to improve the energy resolution without losing detection efficiency for a photon detector used in 4πβ-γ coincidence counting, a study was undertaken to show that new scintillators such as LaBr$_3$:Ce and LaCl$_3$:Ce, activated with cerium (Ce$^{3+}$) atoms as the primary scintillation source, offer an excellent alternative to the NaI scintillator which has the disadvantage of low energy resolution.
List of publications


9. Alexiev, D., Mo, L., Smith, M., “Comparison of LaBr$_3$:Ce, LaCl$_3$:Ce, CZT and NaI(Tl) for Resolution of Nuclear Material Spectra”. Accepted by the *IEEE-9th International Conference on Inorganic Scintillators and their Applications*, June 4-8, 2007, Winston-Salem, USA.

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Chapter One

Introduction

1. General introduction

The development and application of both nuclear power and nuclear medicine have been the major interests in nuclear science for almost half a century. Associated with that are the increasing demands for standards of radioactivity and improved accuracies for radionuclide with a wide variety of decay schemes.

For activity standards, it is not possible to have a physical representation of the Becquerel which is applicable to all radionuclides. As a consequence a standard for any particular radionuclide will only be suitable as a measurement standard for that same radionuclide. This presents considerable difficulty for radioactivity measurements as there are hundreds of different radionuclides, all of which have their own particular decay schemes with different energies, decay rates, intensities and types. Furthermore, there is no one method or detector that can be used for the measurement of all radionuclides. Therefore, standardisation of a particular nuclide involves a particular study of its decay scheme, radiation particle interaction with matter and measurement technique.

The activity measurement can be classified as being either direct or indirect. Direct measurements are the measurements in which the activity is determined from count-rate data alone, without reference to any activity determined from previous experiments. Direct measurements are sometimes referred to as “absolute” measurements, and standards so produced are often designated as “absolute” or “primary” standards. The direct measurements are highly accurate measurements. Indirect measurements are made by performing comparisons with the primary standards through a reference instrument such as an ionisation chamber or gamma ray spectrometer. The standards so produced are often designated as “secondary” standards. “Tertiary” standards are made traceable to secondary standards.

There are variety of absolute methods for activity determination, as described by Allen [Allen 1965] and NCRP [NCRP 1985]. They can be classified into the following categories. The first is defined-solid-angle counting [Bambynek 1967] and 4\pi-counting [Seliger and Schwebel, 1954., Pate and Yaffe 1955a, 1955b, 1955c, 1955d, 1956]. In defined-solid-angle measurements, a point source is placed not too close to the entrance window of a detector and the system is in a vacuum. The detection is made within a defined solid angle much less than 4\pi steradians (\Omega << 4\pi) and the effects of absorption or scattering by either the source itself or its
surroundings are kept minimal and all particles incident upon the sensitive volume of the
detector are recorded. Then the activity can be determined from the count rate, and the solid
angle subtended at the source by the detector. The counters are designed such that the solid
angle is well-defined and directly measurable. When the solid angle increases beyond 10% of
$4\pi$ steradians the defined-solid-angle method becomes unreliable, mainly because the effects of
absorption and scattering are increasing and can no longer be estimated with sufficient
accuracy. Defined-solid-angle counting has been used for measuring alpha-particles with
energies of several MeV, and photons in the energy range of 1 to 80 keV, in another words,
those particles which are “lightly scattered but heavily absorbed” [NCRP, 1985]. For $\beta$-
particles, which result in much more absorption and scattering in the source itself and detector
window, the method is not appropriate.

If the solid angle is increased to $4\pi$ steradians and energy is above threshold, every
radiation particle can be detected, and the count rate is then a direct measure of the
disintegration rate. This eliminates the need for an accurate definition of the solid angle and its
associated scattering problems. The angle need no longer be considered. $4\pi$-counting has been
used for measuring charged particle emitters where the source is made very thin, placed on a
very thin source mount and counted within a windowless detector subtending an angle of $4\pi$
steradians around the source. Forms of $4\pi$ counting, in which the detection efficiency is nearly
100 percent are: $4\pi$ gas proportional counting of a very thin source, internal gas counting of
radionuclides in gaseous form, and liquid-scintillation counting of energetic alpha- and beta-
particle emitters.

These methods are relatively simple in procedure, but they involve some corrections such
as self-absorption, and their accuracies are often limited.

Coincidence counting is the most powerful method for the absolute measurement of
radioactivity. Coincidence counting is used for the absolute radioactivity measurement of a
source when each nuclear event involves two distinguishable radiations being emitted in prompt
succession. Although there are some variations in the method with regards to the choice of
detector type and the selection of radiations detected, the fundamental principle of the
coincidence techniques is the same. The $4\pi\beta-\gamma$ coincidence method is used for radionuclides
that disintegrate by emission of an $\alpha$ or $\beta$ particle, or an X-ray (as a result of electron capture),
followed by a gamma ray in prompt succession. The gamma-gamma coincidence counting is
used for radionuclides that emit two gamma rays in cascade.

The development of new standards is mostly the result of user demand. The effectiveness
and accuracy of a standard relies upon the absolute standardisation techniques and their
subsequent development and dissemination to meet the requirements of the user communities.
The development, maintenance and dissemination of the activity standard are the main task of the national standard laboratory.

2. Review of radioactivity measurement techniques

Following the discovery of X-rays in 1895 by Roentgen [Roentgen, 1895], H. Becquerel discovered radioactivity by accident in February 1896 [Becquerel, 1896a], when using naturally fluorescent minerals to study the properties of X-rays. In Becquerel’s paper, he reported that the double sulphate of uranium and potassium emitted certain rays which gave a reactive impression on a photographic plate even when exposed to no light. In other words, the emission occurred irrespective of whether or not the salt was caused to fluorescence. He realised that the emission came from uranium itself, even in the dark. Radioactivity, which was always present on the Earth, had been discovered. Further experiments showed that uranium minerals were the only phosphorescent minerals to show this effect [Becquerel, 1896b]. On the 7 March 1896, H. Becquerel showed that this radiation had the important property of discharging electrified bodies [Becquerel, 1903]. Later, H. Becquerel reported that this discharge was due to fact that the gas through which the radiation passes is made temporarily to conduct electricity [Becquerel, 1896c]. This discharging power is similar to that of “Roentgen rays”. In November 1896, J. J. Thomson and E. Rutherford theorised that the rays produce positively and negatively charged carriers throughout the volume of the gas surrounding the charged body, and that the rate of production of the charge carriers is proportional the subjected radiation [Thomson and Rutherford, 1896]. In 1899, E. Rutherford published a detailed comparison of the nature of the discharge produced by “uranium radiation” with that produced by “Roentgen rays” [Rutherford, 1899]. The ability of uranium radiation to “ionise” gases formed the basis of a method for the accurate quantitative analysis and comparison of the radiations.

In July 1898, P. and M. Curie [Curie, 1898] discovered polonium which was 60 times more radioactive than uranium. In the same year, M. Curie discovered radium which was 400 times more radioactive than uranium [Currie, 1898]. M. Curie introduced the shorthand term “radioactivite” (radioactivity) to describe the “activite radiante de l’uranium” [Becquerel, 1903]. A year earlier, in 1897, J.J. Thompson, had discovered the electron.

Allisy [Allisy, 1994/95] has given a good review of the discovery of radioactivity and evolution of quantities, units and standards in radioactivity.

In 1899, E. Rutherford identified two types of radiation emitted by uranium minerals, namely the alpha rays, that produced the far greater part of ionisation, but was completely stopped by a thin sheet of paper, and the beta rays, that produced much less ionisation, but were much more penetrating. In 1900, even more highly penetrating radiations emitted from radium,
the gamma rays, were discovered by P. Villard. By successfully deflecting the α particles from radium and its decay products with both magnetic and electric fields, Rutherford showed that the α particles were positively charged. In 1909, Rutherford and Royds [Rutherford and Royds, 1909] first proved that the α particle was an ionized helium atom projected at high speed from a disintegrating atom.

In 1914, J. Chadwick [Chadwick, 1914] first demonstrated that the spectrum of β-radiation was continuous. The continuous spectrum was for a long time completely unexplained, until Fermi [Fermi, 1934] presented a successful quantitative theory of the shape of β-ray spectra and the lifetime of β-ray emitters in 1934. The theory was based on Pauli’s neutrino hypothesis and was quite satisfactorily able to account for the relation between the decay rate and maximum energy of the β-particles. Furthermore, the shape of the spectrum was calculated. The theory was soon extended to all possible types of interactions between heavy and light particles in β-decay. Progress has been more rapid since then, and by the early 1950s many of the fundamental problems concerning β-ray spectra had been solved.

Since Henri Becquerel discovered the radioactivity in 1896, radionuclides have been used in a diverse range of applications within medicine, science and industry and the development of the technique of quantification of radioactive sources has never stopped.

2.1. Early stage of radiation detection

The earliest radiation particle-detection device consisted of a thin layer of zinc sulphide (ZnS) crystal; the scintillations by α-particles striking the ZnS screen were observed. The first devices were only sensitive to the α- and β-particles. In 1908, Rutherford and Geiger [Rutherford and Geiger 1908.] used what they called an “electrical method” to detect α-particles expelled from uranium, thorium, radium, and actinium. They constructed a simple gas-ionisation detector (see Figure 1.) in order to determine by a direct method the number of α-particles expelled from a definite quantity of radioactive matter. The detector consisted of a brass cylinder and a central insulated wire connected to an electrometer. The outer cylinder was charged to a negative potential. The cylinder contained air or gas at a reduced pressure. This detector is considered as a primary innovation in gas counters. Rutherford and Geiger continued developing their technique and by 1912 had increased their average counting rate for α-particles from the 3 to 5 counts per minute to around 1000 counts per minute [Geiger and Rutherford, 1912].
Figure 1. The first detecting vessel built by Rutherford and Geiger in 1908. It consists of a brass cylinder A and a central insulated wire B passing through ebonite cork C at the ends. In the ebonite cork C was fixed a short glass tube D which has a circular opening. The $\alpha$-particles entered the detecting vessel through this opening. A large stop-cock F was attached to one end of the long glass tube E. The other end of the glass tube was closed by a ground stopper G. (Reproduced from [Rutherford and Geiger, 1908])

In 1928, Geiger and Müller [Geiger and Müller, 1928] built a gas-filled detector based on ionisation. It was this type of detector which later became commonly known as the Geiger-Müller counter, or G-M tube. The counter employs gas multiplication to greatly increase the charge generated by the original ion pairs formed along the radiation track. It functions only as a simple counter of radiation-induced events because all pulses from a G-M tube are of the same amplitude regardless of the number of original ion pairs generated. It cannot be used as a radiation spectrometer. Due to the simplicity, low cost, and ease of use, the G-M counter is still widely used.

It was in the same year that Geiger and Klemperer [Geiger and Klemperer, 1928] discovered the proportional region of the gas ionisation which laid the foundations for “proportional counting”. With the proportional property, $\alpha$ and $\beta$-particles could be distinguished by the difference in ionisation produced.

2.2. 4$\pi$ counting of $\alpha$ and $\beta$-particles

Although 4$\pi$-counting was originated in 1944 by Simpson [1944] who described a counter with geometry close to 4$\pi$ steradians, the first real 4$\pi$ counter was reported by Haxel and Houtermans [Haxel and Houtermans, 1948] in 1948. Since then a number of 4$\pi$ counting chambers of a variety of designs have appeared.
In a $4\pi$ counter, the count rate equals to the disintegration rate $N_0$ if absorption in the source or source-mount can be neglected. This is still true when a nuclide emits more than two radiation particles per disintegration, provided the time interval between the two radiations is less than the resolving time of the counter and associated electronics. Thus the system is insensitive to $\gamma$-rays and internal conversion electrons following $\beta$- or $\alpha$- decay. The system is also insensitive to the scattering effect since any discharges subsequent to that corresponding to the primary event caused by repeated scattering of one particle will occur within the counter resolving time. Therefore, $4\pi$-counting eliminates not only geometry correction, but also secondary radiation and scattering effect.

The introduction of the $4\pi$ proportional counter in late 1940s was to meet the requirements of good energy resolution, high detection efficiency, wide-range amplitude response and sufficient short resolving time.

In common with the G-M tube, a $4\pi$ proportional counter is a gas-filled detector, operated in pulse mode and utilizes the phenomenon of gas multiplication to amplify the charge generated by the original ion pairs created within the gas. The difference is that for a proportional counter each original electron leads to an avalanche that is basically independent of all other avalanches formed from other electrons associated with the original ionizing event. Therefore the collected charge is proportional to the number of original electrons, i.e. proportional to particle energies.

$4\pi$ proportional counters were initially operated at atmospheric pressure. Therefore, they are also called “gas flow proportional counter”. These counters are filled with methane gas which flows through the counter at atmospheric pressure. The counters have two plateaus: at the lower voltage for $\alpha$-particles and at higher voltage both for $\alpha$- and $\beta$-particles. The energetic $\beta$-particles deposite only a small fraction of their energies in the counter sensitivity volume. The counters do not give any information concerning the particle energy, but only detect events induced by the particles. ANSTO Ionising Radiation Physics Laboratories employs this type of counter for the $4\pi\beta-\gamma$ coincidence counting.

High pressure was used at a later time and these counters are called “pressurized proportional counter” [Baerg, 1973b, Bek-uzarov, 1973]. The counters are used for X-ray and Auger-electron detection in electron capture nuclide measurements. The pulse height is truly proportional to the energy deposited. These counters are operated under different pressures; they maintain a constant pressure over a long period of measurement. They have small absorption of X- or $\gamma$-rays in the walls of the counter. They have high stability and linearity of amplification factor.
Although a $4\pi$ counter detects all the ionizing radiation emitted from the source, the major problem associated with $4\pi$-counting is source self-absorption of radiation (and on a small scale the absorption of radiation by the source-mount).

The precise evaluation of absorption by the source and source-mount is difficult. Therefore, one should aim to reduce the absorption to as small an amount as possible. The way of reducing source self-absorption is obviously to improve the source preparation technique. One of the methods of source preparation is to evaporate a liquid drop on the source mount. If the area of the source can be kept constant, reduction of the solid content (inactive carrier content) of the drop will reduce the source self-absorption and an extrapolation to zero source weight may be made to determine the activity for zero absorption. However, it is found that evaporation produces aggregates of crystals. Other source preparation techniques, such as using wetting agents like Ludox (colloidal silica) [Merritt et al., 1959], Catanac [Wyllie, 1989] and aluminium treatment [Yoshida et al., 1977] have been investigated. These wetting agents act as nucleating agents in the crystallization process and produce a large number of small source crystals.

The use of VYNS thin film [Pate and Yaffe, 1955a] of a superficial density of 5 - 20 $\mu$gm/cm$^2$ reduces the absorption of radiation in the source mount to a negligible level, although, for the work of the greatest precision and particularly that with the weaker $\beta$ emitters, the absorption loss in the source mount still needs to be accurately evaluated and corrected. The use of very thin films is nevertheless still of considerable advantage, since the magnitude of the correction is reduced, and the final disintegration rate value less sensitive to the errors in the correction.

Despite these improvements in the source and source-mount preparation, the absorption was still of great concern. It was desirable to develop a new direct measurement technique. The $4\pi\beta-\gamma$ coincidence counting technique was then developed to overcome this problem.

### 2.3. Gamma-ray detection

Before considering the $4\pi\beta-\gamma$ coincidence counting, let us review the detection of $\gamma$-rays.

In 1948, Hofstadter [Hofstadter, 1948] first reported the detection of $\gamma$-rays using a sodium iodide crystal to which a trace of thallium iodide had been added in the melt. The crystal produced an exceptionally large scintillation light-output compared with the organic materials that had previously received primary attention. This discovery, more than any others, started the modern era of scintillation spectrometry of gamma radiation. This crystalline scintillator remained for almost 20 years the most important for gamma ray spectrometry. The most extraordinary property of NaI(Tl) is its excellent light yield, giving high detection efficiency for X- and $\gamma$-rays.
The scintillation counter has always been at a basic disadvantage in comparison with the other methods of primary charged particles. Its resolution has kept it in the role of an energy-sensitive detector rather than spectrometer. Even the gas proportional counter is capable of approximately four times better resolution than the scintillation counter (500 eV per photoelectron versus 30 eV per ion pair, where the relative resolution is proportional to $(\text{signal})^{-1/2}$) [Siegbahn, 1965].

Nowadays, scintillation detectors are not considered as high resolution instruments, although they are still widely used for applications that need simplicity of use rather than high energy resolution. However, their high detection efficiency provides great merit for their use in coincidence measurements. Therefore, many laboratories are still using NaI(Tl) in their coincidence counting systems.

In 1949, MacKay [MacKay, 1949] first reported the detection of $\alpha$-particles in the vicinity of the depletion layer of a reverse-biased germanium point-contact diode. However, it was not until 1956, when Mayer and Gossick [Mayer and Gossick, 1956] published results of a study on germanium surface-barrier detectors of a few square millimetres that the development of broad-area junction detectors began.

The first gold-silicon surface-barrier diode detector was described by Davis [Davis, 1958] in 1958. Since then, silicon has been used almost exclusively for charged particle detectors, because high resolution can be obtained at room temperature.

Unfortunately, the low atomic number of silicon ($Z = 14$) makes such detectors unsuitable for $\gamma$-ray spectroscopy. In 1962, the advent of lithium-ion drift compensate germanium ($Z = 32$) technique [Freck and Wakefield, 1962] overcame this problem. Since the atomic stopping power for photoelectric absorption varies approximately as $Z^5$, a Ge detector will be some fifty times more efficient in detection by this process. In 1965, Tavendale [Tavendale, 1965] developed the large volume, high sensitivity coaxial Ge(Li) detector. The Ge(Li) detector became commercially available in the early 1960s and served as the common type of large-volume germanium detector for two decades.

In 1970, very high purity germanium containing $\sim 10^{10}$ electrical impurities per cm$^3$ was successfully grown by Hall et. al. [Hall et al. 1970]. The widespread availability of high-purity germanium in the early 1980s provided an alternative to Ge(Li) and manufacturers have now discontinued production of Ge(Li) detectors in favour of the HPGe type. A major reason for this evolution is the much greater operational convenience afforded by HPGe detectors. Whereas Ge(Li) detectors must be continuously maintained at low temperature, HPGe detectors can be allowed to warm to room temperature between uses. In general, the important performance characteristics, such as detection efficiency and energy resolution, are essentially identical for Ge(Li) and HPGe detectors of the same size.
Current development in γ-ray spectrometry no longer focuses on increasing the energy resolution of a system. Instead, they pursue higher throughputs of signal and higher detection efficiency. High signal throughput is achieved by the improvement of electronic equipment. Higher efficiency is achieved by growing large crystals and improving source detection configurations. While a HPGe detector has been used almost exclusively for γ-ray spectroscopy, it still suffers from limitations in the detection efficiency and the requirement of cooling for optimum resolution.

A detailed review of the development of Ge detectors was given by Alexiev et al. [Alexiev et al., 2002].

2.4. The development of $4\pi$β−γ coincidence counting

2.4.1. Coincidence counting

The coincidence method can be applied to events where the radioactive substance emits several coherent rays in the same disintegration process simultaneously, or in cascades.

In their experiment, Geiger and Werner [Geiger and Werner 1924] in 1924 demonstrated that, with two observers, each using a low-power microscope to record the scintillations resulting from α-particles striking a ZnS(Ag) screen, the efficiency of each of the observers could be determined by the number of counts recorded by each observer, as well as the number of events which were coincident. This was considered as the earliest known application of coincidence counting.

In the late 1920s, electronic components for coincidence were developed [Bothe and Kolhorster, 1929; Rossi, 1931 and 1934; Tuve, 1930]. This resulted almost immediately in the widespread use of coincidence counting in cosmic ray measurements. These measurements were chiefly concerned with the responses of separate Geiger counters to particles and were not associated with the measurement of radioactivity. For example, in Bothe and Bayer’s arrangement [Bothe and v. Baeyer 1935], the source was placed inside the anode of a G-M counter for detecting the first particle, and this counter was surrounded by a ring of counters connected in parallel, forming the second counter of the coincidence equipment for detecting second particles. The solid angle subtended from source to counter for both rays was practically equal to $4\pi$.

In 1940, the coincidence method was first applied to the direct measurements of radioactivity by Dunworth [Dunworth, 1940]. The possible errors due to the angular correlation of coherent rays, gamma-gamma coincidences in the beta-gamma coincidence method, scattered
gamma-rays, fluctuation of the resolving time and fluctuations of the background were reviewed by Barnothy and Forro [Barnothy and Forro, 1951].

Subsequently, the $\beta-\gamma$ coincidence method, independent of detection efficiency, became a powerful technique for simple $\beta-\gamma$ emitting radionuclides provided that either the $\beta$-counter or the $\gamma$-counter was equally sensitive to all parts of the source [Putman, 1950]. This could be achieved by careful choice of detectors and preparation of uniform sources. Putman [Putman, 1957] later suggested that the uniformity of specific activity is not a necessary condition, but at least one detector should have the uniform sensitivity over the entire source. This condition is closely approximated in practice for the $\gamma$-detector. The $\beta-\gamma$ coincidence method was particularly useful for determining the self-absorption correction in $4\pi\beta$ counting and for determining the efficiency of radiation detectors.

In principle, the coincidence method may be applied to any known decay scheme. However, the convenience and accuracy of the method decreases rapidly as the complexity of the decay scheme increases [Snell, 1962]. For these reasons, in the early days, the coincidence method was generally only used for measuring radionuclides that disintegrate with the emission of a single $\beta$-particles followed virtually immediately by one or more $\gamma$-rays.

In practice, there are a number of corrections that need to be applied, such as those for source size, angular correlation, internal conversion, dead-time, sensitivity of the $\beta$-detector to $\gamma$-photons, accidental coincidences and background fluctuations.

2.4.2. $4\pi\beta-\gamma$ coincidence counting

The $4\pi\beta-\gamma$ coincidence counting method is an improvement of the ordinary $\beta-\gamma$ coincidence method. The first and well known paper which described the $4\pi\beta-\gamma$ coincidence counting was written by Campion [Campion 1959]. Campion demonstrated that by using the $\beta$-detector which has $4\pi$ geometry (high $\beta$- detection efficiency) for the coincidence method, most of these corrections are small and hence the accurate measurement of $\beta-\gamma$ emitting nuclides can be achieved. A block diagram of Campion’s $4\pi\beta-\gamma$ coincidence counting system is presented in Figure 2.
The 4πβ−γ coincidence counting technique can be also used for the measurements of electron capture nuclides, where the measured beta efficiency is that of the Auger electrons and X-rays associated with the electron capture process. The original 4πβ−γ coincidence counting and its extended method have been used extensively for the standardization of wide range of radionuclides [Campion, 1961].

The conventional 4πβ−γ coincidence equation, \( \frac{N_\beta N_\gamma}{N_c} = N_0 \), applies to a point source with a simple decay scheme (a single β emission followed by one or more γ-rays). \( N_\beta \), \( N_\gamma \) and \( N_c \) are the count rates measured by the β-detector, γ-detector and coincidence recorder, and \( N_0 \) is the disintegration rate. The β-detector is assumed to respond only to β-particles, the γ-detector only to γ-rays and the coincidence recorder only to simultaneous pulses from the two detectors (true coincidences).

In any practical system, however, at least one of the detectors, usually the β-detector, is not exclusively sensitive to one type of radiation, i.e. β-particles only, but to γ-rays and conversion electrons as well. Therefore, the more general and sophisticated coincidence equation [Baerg, 1966] has been developed and can be expressed as \( \frac{N_\beta N_\gamma}{N_c} = N_0[1+K] \), where \( K \) is the decay-scheme-dependent correction which corrects the influences of those unwanted radiations, including the γ-sensitivity of the β-detector, the detection of internal conversion electrons and different β-branches.
2.4.2.1. $4\pi\beta-\gamma$ coincidence extrapolation method

To eliminate these influences, the most practical technique is the $4\pi\beta-\gamma$ coincidence extrapolation method [Williams and Campion, 1963. Baerg, 1966 and 1973, Houtermans and Miguel, 1962]. This method requires no knowledge of the decay scheme parameters and detection efficiency, but only the observed count rate. It has proven to be the most accurate method for activity determination. However, to be able to achieve the highest accuracy with the method, the highest attainable beta detection efficiency needs to be more than 90% so that the extrapolation range can be made as short as possible. Repeating measurements under different experimental conditions is required.

With the efficiency extrapolation method, a source is measured with various $\beta$-particle detection efficiencies. The apparent disintegration rate $N_\beta N_\gamma / N_c$ is plotted against $(1-\varepsilon_\beta) / \varepsilon_\beta$. Extrapolation of $N_\beta N_\gamma / N_c$ to $\varepsilon_\beta = 1$ yields the $4\pi$ beta count rate to be $N_\beta = N_0$ and the slope to be the correction. The relationship between the apparent disintegration rate and $(1-\varepsilon_\beta) / \varepsilon_\beta$ is often referred as the efficiency function. The efficiency extrapolation method is based on the assumption of that all the branches are subject to the same physical condition which causes the counting losses, so the inefficiencies of the various $\beta$-branches have a linear relationship. The extrapolation method eliminates the influence of conversion electrons and gamma sensitivity of the $\beta$ detector.

Experimentally, $\beta$-particle detection efficiencies can be varied by 1) changing source self-absorption, 2) adding thin absorbers, and 3) adjust electronic discrimination levels. Methods 2) and 3) are commonly used. Method 3) is used when the $4\pi\beta$-counter is pressurised. The three acceptable methods of efficiency variation are not theoretically equivalent [Baerg, 1966 & 1967a], although experimentally the differences are insignificant [Baerg, 1967b].

The efficiency extrapolation method was first used by Steyn and Haasbroek [Steyn and Haasbroek 1958] and subsequently developed by Campion et. al. [Campion et. al., 1960], Taylor [Taylor, 1962], Taylor and Merritt [Taylor and Merritt, 1962], Williams and Campion [Williams and Campion, 1963]. The method is now widely used for the measurements of radionuclides with complex decay schemes.

2.4.2.2. Theoretical correction

The decay-scheme-dependent correction $K$ can be also derived theoretically by consideration and calculation of all elementary processes in the source, although some parameters in general have still to be determined experimentally. This method requires
knowledge of decay scheme parameters and subsidiary information on some detector efficiencies. This method is based on a single measurement and has advantage of avoiding lengthy measurement procedures. When beta efficiency is low, for example, in the case of measuring a thick source, this becomes the only method to be adopted.

For the correction \( K \), \( \gamma \)-sensitivity \( \varepsilon_{\beta\gamma} \) of the \( \beta \)-detector is the most difficult parameter to determine. The original method given by Campion [Campion, 1959] for determining \( \varepsilon_{\beta\gamma} \) was to count sources of known activity covered with absorbers, usually polyethylene, thick enough to absorb all the \( \beta \)-particles, and it was assumed that all the counts observed in the \( \beta \)-detector were due to the \( \gamma \)-sensitivity of the \( \beta \)-detector. Campion measured \(^{141}\)Ce, \(^{203}\)Hg, \(^{95}\)Nb and \(^{60}\)Co. \( \varepsilon_{\beta\gamma} \) vs \( \gamma \)-ray energy was plotted. \( \varepsilon_{\beta\gamma} \) was found to be in the range of approximately 0.3 – 1.1% for an energy from about 200 keV to 1332 keV. However, the thickness of the absorbers required to stop all the \( \beta \)-particles is so large that the \( \gamma \)-ray interactions in the foils cannot be neglected and much higher results are obtained due to photoelectron and Compton electrons ejected into the gas from the foils. It is difficult to separate this effect from the inherent \( \gamma \)-ray sensitivity of the detector.

In order to overcome this problem, Williams and Campion [Williams and Campion, 1963] used the 4\( \pi \)\( \beta \)–\( \gamma \) coincidence counting efficiency extrapolation method to obtain much more accurate values of \( \varepsilon_{\beta\gamma} \) as a function of \( \gamma \)-ray energy. With this method, \( \varepsilon_{\beta\gamma} \) was obtained from the slope of the extrapolation function. A function of \( \varepsilon_{\beta\gamma} \) vs \( \gamma \)-ray energy was plotted by measuring \(^{51}\)Cr (\( E_\gamma = 320 \text{ keV} \)), \(^{95}\)Nb (765 keV), \(^{54}\)Mn (835 keV), \(^{60}\)Co (1332 keV) and \(^{88}\)Y (1836 keV). \( \varepsilon_{\beta\gamma} \) was found to be in the range of approximately 0.1 – 0.5% for the energy about 320 keV – 1836 keV. The accuracy of this method was later confirmed by Merrit and Taylor (Merrit and Taylor, 1966). The method may offer much improved results compared with the original absorber method, but the use of thin absorbers to vary beta efficiency may still cause unexpected photon interactions in the absorption foils. The range of absorber thicknesses (and hence efficiencies) that can be used is limited by the requirement that they stop a negligible number of \( \gamma \)-rays; otherwise the secondary electrons produced will increase the apparent \( \varepsilon_{\beta\gamma} \). The detection efficiency for conversion electrons may be reduced if considerably thick absorbers are used.

Kawada [Kawada et al., 1968, Kawada, 1969] developed a method in which the \( \beta \)-efficiency was varied by changing the counter geometry from 4\( \pi \) to 2\( \pi \), using the top- and bottom-halves of the 4\( \pi \) counter. The results were found to be similar to those of Williams and Campion, and Merritt and Taylor. Since this method requires no additional absorber foil and the measurements can be made without changing any physical parameter, precise results can be expected. However, this method requires modification of the instrument.
There is an additional contribution to the coincidence counting rate, which arises from Compton scattering or, in more complex decay schemes, from the presence of other cascade $\gamma$-rays ($\gamma-\gamma$ coincidences). This effect can be reduced to negligible levels by appropriate energy discrimination in the $\gamma$-channel (setting a window for the $\gamma$-photopeak).

Regarding the detection efficiency of the $\beta$-counter for conversion electrons in a thin source, the efficiency for energetic conversion electrons is taken as unity in $4\pi\beta-\gamma$-counter geometry [Campion, 1959] unless they are of extremely low energy (< 50 keV). However, for conversion electron efficiency less than unity, the event may still be recordable by detection of X-rays or Auger electrons.

Perolat [Perolat, 1973] pointed out that the accuracies of the two methods, extrapolation and correction, are more or less equal for the simple $\beta-\gamma$ decay scheme radionuclides (e.g. $^{60}$Co, $^{46}$Sc, $^{56}$Mn and $^{95}$Nb). For complex decay schemes, the best accuracies are obtained by the extrapolation method (e.g. $^{133}$Ba, $^{105}$Au, $^{106}$Ru-Rh). Nevertheless, in many cases, the theoretical correction method can provide satisfactory accuracy, although it may not be as good as the extrapolation method. Some examples of using the theoretical correction method are the measurement of $^{65}$Zn, $^{99}$Mo and $^{59}$Fe.

2.4.2.3. $4\pi\beta-\gamma$ coincidence efficiency-tracing technique

The $4\pi\beta-\gamma$ coincidence counting technique has been also extended to pure $\beta$-emitters by addition of suitable $\beta-\gamma$ emitters as the tracer of the counting efficiency, the so-called efficiency-tracing technique [Campion et al., 1960, Baerg etc., 1964, Steyn, 1973, Lowenthal, 1973]. The method involves combining into a single counting source the pure $\beta$-emitter and an accurately known fraction of a $\beta-\gamma$ emitting “tracer”. This has been achieved by (1) combining atoms of the two different nuclides into the one molecule [Campion and Merritt, 1960], and (2) quantitatively mixing a solution of the pure $\beta$-emitter with a solution of the tracer [Campion and Merritt, 1960]. The tracer must have a simple decay scheme which is “not-too-dissimilar” to that of the pure $\beta$-emitter.

The total count rate of the mixture is found by extrapolating to 100% efficiency. The activity concentration of the nuclide being traced is obtained by subtracting the count rate due to the tracer. In fact, the tracer is used to determine the $\beta$-detection efficiency of the source.

Ideally the shape and end-point of the $\beta$-spectra of the pure beta emitter and the tracer nuclide should be the same. However, in practice this is often difficult to achieve. The effects of the shape of the $\beta$ spectra of $\beta-\gamma$ tracers and pure $\beta$ emitters are recognised, and accepted,
though not without reservations [Bryant et al., 1967]. The spectrum effect was investigated by Lowenthal [Lowenthal, 1973]. It was found that measurements made with different $\beta-\gamma$ tracers resulted in different results on extrapolation due to the shapes of the $\beta$ spectra affecting the efficiency tracing function. If the low-energy ends of the spectra are similar in shape and their end-point energies do not differ too much from each other the uncertainties are negligibly small. If the shapes of the two spectra are similar but the difference between the end-point energies is large, the uncertainties are greater. The effect could be even more serious when the shapes of low-energy sections of the spectra differ markedly.

However, the study shows that there is linear relationship of counting efficiencies among many $\beta$ emitters when homogenously mixed: for example, using $^{134}$Cs as a tracer for $^{63}$Ni, $^{60}$Co for $^{35}$S, $^{185}$W [Chun et al. 2002]. For a thin mixed source (achieved by using a wetting agent such as Catanac) whose nuclides are chemically uncombined but homogenously mixed, if the $\beta$ efficiency is varied by using an external absorber, a linear plot of efficiency is often obtained. In order to avoid any differential effects due to separation of the two activities during crystallization as the source is dried, it is necessary to incorporate the two activities into the same chemical compound. $^{82}$Br, $^{24}$Na, $^{177}$Lu, $^{60}$Co and $^{134}$Cs have been used as tracers [Campion, etc., 1960]

2.4.2.4. Some additional points for 4$\pi$$\beta-\gamma$ coincidence counting

In order to maintain the linear functional relations between the inefficiency of different beta branch particles, the $\beta$-detection must be arranged so that 1) beta efficiencies for all branches must be single-valued functions of the variable efficiency parameter $N_\beta/N_\gamma$, and 2) the relating functions must approach to unit detection efficiency for all branches simultaneously [Baerg, 1966]. This requires that the $\beta$-detection geometry be 4$\pi$ steradians. The source mounting arrangement, even in a 4$\pi$ gas proportional counter, can reduce the effective geometry [Baerg, et. al., 1964]. In addition, the counter should be sensitive to 25 ion pairs or less (i.e. $\approx$ 1 keV equivalent energy). An adequate approximate measure of the sensitivity can be obtained using a low Z electron capture source. A 4$\pi$ proportional counter must be also sufficiently sensitive to high $\beta$-energies, since linear energy transfer for $\beta$-particles decreases with increasing energy up to about 1 MeV and the number of ion pairs generated in a practical counter at atmospheric pressure may not exceed 100 for energetic particles.

The 4$\pi$ proportional counter is usually made of aluminium, and is of the pill-box type, with a sensitive volume 3.8 - 5.9 cm in diameter and 5.1 - 7.8 cm in height. It has zero intrinsic noise, low sensitivity to $\gamma$-radiation and a recovery time of the order of 100 ns.
Sodium iodide scintillation counters are currently the most satisfactory for the $\gamma$-detection, providing generally adequate energy resolution. Some laboratories have shown interest in the HPGe detector which provides much better resolution. LaCl$_3$ and LaBr$_3$ detectors have high efficiency and high resolution and could be promising alternatives for $\gamma$-detection in $4\pi\beta-\gamma$ coincidence counting.

### 2.4.3. Digital coincidence counting and other studies

The principles of the $4\pi\beta-\gamma$ coincidence counting method, as well as the nature of the corrections required, are well understood. However, valid application of these principles to actual measurements is not simple and efforts on the improvement of the accuracy of $4\pi\beta-\gamma$ coincidence counting have continued for decades.

The accurate fulfilment of the linearity conditions of the gamma-ray channel was examined by Grigorescu [Grigorescu, 1982] and was found to be crucial for elimination of the error from source self-absorption and the error due to absorption in source mount. Having investigating $^{59}$Fe and $^{134}$Cs, Miyahara and Watanabe [Miyahara and Watanabe 1984] found that to obtain the highest beta efficiency, the crystallites in the $4\pi\beta$ source must be as small and uniform as possible.

In the extrapolation method, a linear function is usually used as the efficiency function; a polynomial function of second or third order is used occasionally. However, if the attainable beta efficiency is not high enough, the first-order fitting function may introduce systematic errors. This problem can be overcome by fitting a polynomial function joining both first and second order terms [Miyahara et al., 1986].

Other developments in the coincidence technique include replacing the $4\pi$ proportional counter with the surface barrier silicon detector for $\alpha-\gamma$ emitters [Koskinas and Dias, 1989] and using NaI crystal detectors to perform $\gamma-\gamma$ coincidence counting for a nuclide emitting two $\gamma$-rays in cascade [Brinkman et al., 1963a and 1963b]. With the $\gamma-\gamma$ coincidence technique the activity is computed from the area $A_1$ and $A_2$ of the photopeaks, the area $A_{12}$ of the sum-peak, and an extrapolation of the total count-rate to zero discrimination. The coincidence equation for this technique is $N = \frac{A_1 A_2}{A_{12}} + T$, where $N$ is the activity of the source, $A_1$ and $A_2$ are the respective areas under the photopeaks of $\gamma_1$ and $\gamma_2$, $A_{12}$ is the area under the sum-peak and $T$ is the area under the whole spectrum. This method has been used to directly measure the activity of $^{125}$I [Simpson and Meyer, 1989].
For decades, the conventional methods of data acquisition for the $4\pi\beta-\gamma$ coincidence counting technique have used dedicated analogue electronic modules such as single channel analysers, gate and delay generators, paralysis units, time-to-amplitude converters, coincidence mixers and multi-channel analysers. The high cost of procurement and maintenance of such systems and the considerable equipment set-up effort have emphasised the need for a more flexible and robust data collection process. Digital coincidence counting is a promising new approach to the absolute measurement of radioactivity. Buckman and Ius published the first paper [Buckman, 1995, Buckman and Ius, 1996] introducing a digital coincidence counting system into $4\pi\beta-\gamma$ coincidence counting. The paper described a digital coincidence-counting system which comprises a custom-built data acquisition card and associated PC software. Using high-speed analogue-to-digital converters (ADCs), the pulse-trains from two radiation detectors are digitised and recorded. Processing of the pulse-trains is performed by a series of software components. The method has the advantage that once a set of pulse-trains has been recorded, it may be processed any number of times by a variety of methods. It avoids the complex and time consuming set up procedures of a conventional analogue system. The software of the system was improved afterwards and the system was verified with the $^{60}$Co, $^{22}$Na and $^{153}$Sm [Buckman et. al, 1998, Butcher et al, 2000]. Good results have been achieved.

2.5. **Liquid Scintillation TDCR and CIEMAT/NIST methods**

2.5.1. **Early stage of liquid scintillation counting**

Liquid scintillation counting was used for relative activity measurements of $\beta$-emitters at an early time, and $\alpha$-emitters later. In this technique the active solution is mixed with a solution of liquid scintillator. As $\alpha$- or $\beta$-particles are emitted, the scintillator emits photons which can be easily detected and counted. In this way the source self-absorption is avoided. Similarly, effects of wall scattering are eliminated because the detector is effectively of $4\pi$ geometry. However, particles emitted near the wall of the scintillator container, travelling towards the wall may not expend all their energy in the scintillator. This will give rise to an excess of low energy pulses and corresponding reduction in the number of high energy pulses - the so-called “wall effect”.

When liquid-scintillation counting was first introduced in the beginning of 1950, the detection efficiency of liquid scintillation counting was very low with only a few percent for tritium due to the instability of the solutions and low quantum efficiency. The early system consisted of a single phototube. For counting a source with low energy and low activity, the
background counting rate in the phototube was a severe limitation. An improvement was made by using a pair of phototubes facing a common scintillation source. Because there are multiple photons emitting in the sample, it is likely that both phototubes will detect the light and thus the utilization of coincidence technique is possible. The randomly occurring background events (thermal noise) in one phototube are only infrequently coincident with those in the second tube, whereas radiations from the source may generate coincidence events between two tubes. Therefore, a high degree of discrimination against background counts is feasible. Systems operating with two phototubes in the coincidence mode have a reduced efficiency but avoid the effects of thermal noise and afterpulsing [Smith, 1980].

A two-phototube system does not give enough information to determine the experimental detection efficiency without an additional reference standard or tracer, and therefore is used for indirect activity measurement. The tracer technique relied either upon quench curves or compensating for the quenched state. Various procedures have been employed, such as extrapolation of the integral counting rate vs discrimination level [Flynn et al, 1971, Jones and McNair, 1970, Houtermans, 1973] and efficiency tracing [Grau Malonda and Garcia-Torano, 1982, Grau Malonda, 1982, Coursey et al, 1984, Coursey et al, 1986].

2.5.2. The development of TDCR method

The introduction of a third phototube into the system, arranged to also detect double coincident scintillations as well as triple coincidence, from the source made it possible to directly measure the activity. Thus, both double- and triple-coincidence counting rates are acquired and the so called “triple to double coincidence ratio (TDCR)” method is used to calculate the detection efficiency. The TDCR method is direct in that it is non-comparative, although it does rely on decay scheme parameters to calculate the two- and three-phototube efficiencies. The advent of phototubes with high quantum efficiencies has permitted the practical application of this system. The TDCR method has become the preferred method for the absolute activity measurement of α- and β-emitters, and has been extended to electron-capture radionuclides and beta-gamma emitters.

Broda [Broda, 2003] has given an excellent review on the development of TDCR method. An overview of his paper is presented as follows.

The first three-phototube system [Figure 3a] was constructed by Hoegl and Schwerdtel [Hoegl and Schwerdtel, 1963] in 1963. However this system was used for relative measurements of pure β-emitters [Schwerdtel, 1966a, b]. In their experiment, the counting efficiency $\phi$, was determined using a set of quenched standards. Counting rates of the triple ($N_T$) and double ($N_{ab}$) coincidences were measured between three phototubes and a simple pair of
phototubes, respectively. The experimental ratio \( K = \frac{N_T}{N_{ab}} \) was calculated. The function \( \phi(K) \) was used to determine the activity of the measured sample.

In 1970, Kolarov et al. [Kolarov et al., 1970] suggested an absolute method of determining the activity of \( \beta \)-emitters based on a simple detection efficiency model using two phototubes working in sum and in coincidence. The major disadvantages of this method were the effect of afterpulses and thermal emission noise observed in the sum channel.

The above triple coincidence LS detector and the principle of the detection efficiency model have both been the basis of the TDCR absolute method of radionuclide activity determination.

The original version of the TDCR method was presented by Pochwalski and Radoszewski (Pochwalski and Radoszewski, 1979) in 1979. In Pochwalski’s system (Fig. 3b), the outputs of three double coincidence gates \( (N_T) \) and the sum of double coincidences \( (N_D) \) (including the triple coincidences) were obtained. Note that they are different from Schwerdtel’s system. The experimentally determined TDCR value, denoted by \( K \), is given by

\[
K = \frac{N_T}{N_D}.
\]

Obviously \( 0 \leq K \leq 1 \), since the triple coincidence-counting rate, \( N_T \), is always lower than the double coincidence-counting rate, \( N_D \). As the counting efficiency approaches unity, the double- and triple- coincidence-counting rates will approach the disintegration rate of the source \( N_T \to N_0 \), \( N_D \to N_0 \) and the ratio \( K \) will also approach unity \( K \to 1 \). By extrapolating to \( K \to 1 \), the values of the disintegration rate, \( N_0 \), can be calculated. Thus the ratio \( K \) was found to be a very convenient experimental indicator of the counting efficiency of a given sample. In order to obtain series of pairs of count rates \( N_T \) and \( N_D \), the efficiencies were varied by changing the optical efficiency or quenching state. Pochwalski placed a cylindrical steel wire spring around the counting vial. The number of coils appearing between the vial and photocathodes changed with the pressure applied on the spring and hence the optical efficiency of the phototubes varied.
The principle of the detection efficiency model applied to the TDCR method is based on the detection efficiency model for the two phototubes system elaborated by Kolarov et al [Kolarov et al. 1970]. Theoretical calculations of counting efficiency and the comparison with the experimental data for pure $\beta$-emitters for TDCR method using this model were first reported by Broda et al [Broda et al 1988]. This model assumed that the three phototubes had the same quantum efficiency.

In the early 1990s, the electronics of the TDCR system was modified to introduce three additional double coincidence counting outputs $N_{ab}$, $N_{bc}$ and $N_{ac}$ [Vatin, 1991, Cassette and Vatin, 1992, Broda and Pochwalski, 1992] in addition to the two shown in Figure 3 (b). In this enhanced system, the electronic coincidence unit gave five outputs: three double coincidences, AB, BC and CA, one triple coincidence ABC and a logic sum of the three double coincidences including triple coincidences, AB+BC+CA, as shown in Figure 4. An extendible dead-time generator was incorporated in the electronic system. This type of dead-time eliminates spurious pulses caused by either phosphorescence or the residual gas in the phototube. With this system, four equations, written as below, with counting efficiencies ($\phi_{ab}$, $\phi_{bc}$, $\phi_{ac}$ and $\phi_T$) from four coincidence channels, could be established, and the activity $N_0$ could be determined.
\[ N_{ab} = N_0 \phi_{ab}(\varepsilon_a, \varepsilon_b), \]
\[ N_{bc} = N_0 \phi_{bc}(\varepsilon_b, \varepsilon_c), \]
\[ N_{ac} = N_0 \phi_{ac}(\varepsilon_a, \varepsilon_c), \]
\[ N_T = N_0 \phi_T(\varepsilon_a, \varepsilon_b, \varepsilon_c). \]

Figure 4. Block diagram of the TDCR coincidence unit (reproduced from reference [Broda and Pochwalski, 1992])

In this way, the efficiencies of the individual phototubes, \( \varepsilon_a, \varepsilon_b \) and \( \varepsilon_c \), are directly determined. Thus the assumption of symmetrical behaviour of the three phototubes is no longer necessary. In fact, the real asymmetry can only be ignored for high counting efficiency. Errors introduced by the symmetry simplification increase when the efficiency decreases [Grau Malonda and Coursey, 1988].

In the late 1990s, an electronic module MAC3 (module d’acquisition de coincidences triples) was developed to process the pulses delivered by the three phototubes [Bouchard and Cassette, 2000]. This module, replaces the complex interconnection of nine independent NIM electronic instruments. The module is illustrated in Figure 5.
Figure 5. A simplified block-diagram of MAC3 module. An incoming pulse will set memory (MA, MB, and MC) and, after a delay (B40 ns), will trigger the dead-time circuit. During this delay (coincidence resolving time), it is possible for additional pulses from a different channel to also be registered. After that delay, the system will not accept additional pulses until the (extendible) dead-time period has elapsed. Memories are sampled by a strobe. An arrangement of gates and buffers allows the recording of the counts in each channel. (Reproduced from reference [Broda, 2003])

Determination of the activity of a source by the TDCR method involves best estimation of the ionisation quenching parameter kB value [Birks, 1951]. This is carried out by performing a series of measurements of the source at different quenching levels. The kB value describes the non-linearity of the scintillator. The best kB value should give constant activity over different detection efficiencies. There are several methods of varying the detection efficiency: chemical quenching, use of coaxial grey filters and defocusing of photomultiplier [PM] tubes by reducing the focusing electrode potential. The first two processes reduce the mean quantity of light emission. The last reduces the detection probability. Cassette et al. [Cassette et al., 2000] indicated that the three procedures were equivalent. However, defocusing is more practical than the other two processes.

The TDCR device has also been applied to $4\pi\beta$ (LS)-$\gamma$ coincidence counting, where the three phototube detector is used in the beta channel and the HPGe detector in the gamma channel [Cassette et al., 1994, Bobin and Bouchard, 2006]. With this arrangement the TDCR method can be extended to a nuclide having $\beta-\gamma$ and EC-$\gamma$ decay-scheme. The system also allow a direct comparison between TDCR and the $4\pi\beta$ (LS)-$\gamma$ coincidence method. Bobin and
Bouchard [Bobin and Bouchard, 2006] published the preliminary results of standardising electron capture nuclides $^{54}$Mn and $^{65}$Zn using the TDCR incorporated $4\pi\beta$ (LS)-$\gamma$ coincidence.

2.5.3. The CIEMAT/NIST method

Another widely used method in liquid scintillation counting is the CIEMAT/NIST efficiency tracing method, named after the institutes, Centro de Investigaciones Energeticas Medioambientales y Tecnologicas, Spain and National Institute of Standards and Technology, US, that originated it. The system involved is a two-phototube system and commercially available. Although not a direct activity measurement method, it allows relative standard uncertainties of less than 1% to be achieved even for low-energy $\beta$-emitters ($E_{\beta_{\text{max}}} > 50$ keV).

The CIEMAT/NIST method originated in 1982, when Grau Malonda and Garcia-Torano [Grau Malonda and Garcia-Torano, 1982] presented a method for the calculation of counting efficiency for $\beta$-ray emitting nuclides in a two-phototube system. The author used a set of tritium standards to obtain an experimental quenching curve, from which the $\beta$-particle counting efficiency was calculated. Tritium is a low-energy $\beta$-ray emitter. Due to the low energy, the uncertainty in tritium standard has little influence on the calculated efficiency of the more energetic $\beta$-ray emitters. In the same year, the author extended this method to electron capture nuclides, which are more complicated to measure than $\beta$-ray emitters [Grau Malonda, 1982]. Different ways of atomic rearrangement and their probabilities have to be considered. A Monte-Carlo simulation had to be used for computing the escape probability of an X-ray.

In 1986, Coursey et al. [Coursey et al., 1986] from NIST and CIEMAT elaborated this method with more detailed processes and studies. Since then, it has been widely used for standardizing $\beta$-ray emitting radionuclides.

3. Maintenance, dissemination and application of Activity standards

3.1. Maintenance of activity standards

Unlike other physical quantities (such as mass), an activity standard is not permanent; it decays as time goes by. So once the primary standard is realized for a certain radionuclide, measurements must be taken to maintain the result of the realisation procedure by calibrating a suitable secondary instrument against the primary standard, unless the half-life is sufficiently long and the standard itself sufficiently stable. Calibration of a suitable secondary instrument also avoids using the time-consuming primary method for routine activity measurements.
The re-entrant type, pressurized $4\pi\gamma$ ionisation chamber, coupled with a pA range electrometer is a very stable detector. It is simple to use and the measurements are precise and reproducible. It is therefore an ideal system for use as working standard for $\gamma$-ray emitters. The instrument is normally calibrated for a variety of radionuclides, based on direct measurements. The unit of activity is maintained by radionuclide specific calibration factors for this ionization chamber, expressed as pA/MBq. On the basis of these calibrations, a curve of the ionisation chamber response as a function of $\gamma$-energy is determined. When directly determined factors are not available, this curve can be used to indirectly determine calibration factors for $\gamma$-emitting nuclides by interpolation using the emission probability and energies of the $\gamma$-rays of the radionuclides of interest. A major problem in determining an efficiency curve is that there are only a few monoenergetic photon-emitting radionuclides with the appropriate energy which allow the photon efficiency to be directly calculated, for example, $^{51}$Cr, $^{85}$Sr, $^{54}$Mn and $^{60}$Co (weighted mean energy). For further points, an iterative procedure is usually necessary. One strong photon component of a radionuclide with a more complex decay scheme is used. The weaker components from the measured radionuclide efficiency is subtracted after having interpolated the corresponding photon efficiencies in the known part of the curve and multiplying these by the corresponding emission probabilities.

Assuming the charge collection in the ionisation chamber is 100%, the mean energy deposited in the ionised gas is related to the calibration factor by the relationship $C = \frac{e}{W} E_d$, where $C$ (coulomb per decay) is the calibration factor for a given radionuclide undergoing gamma decay, $e$ (coulomb) is the electron charge, $W$ (eV) is the mean energy absorbed in the gas per ion pair formed and $E_d$ (eV) is the total mean energy deposited in the gas volume per decay. $E_d$ is calculated by $E_d = \sum_{k=1}^{n} E_k I_k$, with $E_k$ (eV) the energy deposited per emitted photon for the $k$th $\gamma$-ray and $I_k$ the emission probability.

In establishing the response function, the contribution from bremsstrahlung photons produced by the slowing down of the beta particles in the surrounding media may have to be included if the nuclide emits high energy beta rays. It is often difficult to define explicitly a detector’s sensitivity to beta rays because they have a continuous energy spectrum and can be absorbed strongly by surrounding media. Similar to the method used for the response function of photon energies, the ionisation chamber response function of beta average energies ($E_{avg}$) can be established by measuring pure beta emitters such as $^{204}$Tl, $^{85}$Kr, $^{89}$Sr, $^{32}$P, $^{90}$Sr, $^{144}$Ce, $^{106}$Ru [Michotte, 2002]. These are the only available radionuclides for this purpose and the function established in this way is much less accurate than that for $\gamma$-ray emitters.
An ionisation chamber itself does not have energy-discrimination capability. The presence of radioactive impurities has to be corrected in conjunction with $\gamma$- or $\beta$-spectrometry measurement. The ionisation chamber response functions of photon and $\beta$-ray average energies are used to evaluate the corrections for any $\gamma$- or $\beta$- impurity when the calibration factor of this impurity is not known.

The stability is continuously checked with stable sources of long-lived radionuclides such as $^{226}$Ra, $^{94}$Nb or $^{166m}$Ho. The use of long-lived check sources can eliminate the inaccuracies and instability of the system.


With regards to maintenance of the activity standards, in principle any other radiation measuring instrument, such as Na(Tl) $\gamma$-counter, HPGe detector and Cerenkov counter etc, could be used for the maintenance of the activity standard, but with regard to both stability and simplicity, none of them can compete with the ionisation chamber. Alternatives have to be found for pure $\alpha$- and $\beta$-emitters.

3.2. Dissemination of the activity standards

The most practical way to disseminate radioactivity standards is using solid sources or solutions. Once the activity concentration of the master solution is obtained as the result of the standardisation procedure, the activity or activity concentration of all sources quantitatively produced from it by weighing and dilution can be used for distribution as secondary standards. The national standards laboratories usually store such standards, e.g. $^{60}$Co, $^{137}$Cs, $^{152}$Eu. The relative uncertainty of the activity or activity concentration is larger than that of the primary standards due to the introduction of additional components in the calculation e.g. weighing error, instability correction and linearity deviation in the low and high pA range of the ionisation chamber.

Nuclear medicine has seen rapid growth in the past two decades of diagnostic and therapeutic treatments of a variety of diseases, including coronary heart disease, chronic lung infections, bone and joint disorders, psychiatric and neurological conditions, and various types of cancer. All of these applications require an accurate measurement of the injected radiopharmaceutical. As with most therapeutic procedures, there is an optimum range of treatment dose. Doses below the range run the risk of incomplete removal of diseased tissue, whilst doses in excess of the range may produce secondary adverse effects in normal tissue. The regulations of many countries require, in most radiotherapeutic application, that the injected activity must be within +/- 10% of the prescribed dosage. This implies, therefore, that those who
produce, supply or use radioactive materials for nuclear medicine must be able to make accurate activity measurements.

Australian radiopharmaceutical producers use pressurised ionisation chambers to routinely measure the activity of radionuclides, and γ-ray spectrometers to identify the presence of impurities and to determine their relative levels. Both types of detector must be calibrated using standard sources. Proportional counters are used to detect the presence of α- or β-emitting impurities. Nuclear medicine departments in hospitals use a radionuclide calibrator (commonly Capintac dose calibrator). These calibrators typically consist of a well-type ionisation chamber, a high-voltage power supply and an electrometer. The electrometer and power supply are normally housed in the same unit. A digital display indicates the activity of the sample.

Both ionisation chambers and dose calibrators are sensitive to photon energy, source geometry and density of solution. If there is any change in any of these parameters, new calibration factors need to be derived.

Manufacturers of calibrators normally provide a set of calibration factors for commonly encountered nuclides. However, these factors will normally be for geometries different to those being routinely used. Furthermore, in order to be traceable to the national standards of measurement, the chamber should be calibrated using appropriate standard sources which are legally traceable.

Investigation of dial settings of Capintec dose calibrators have been performed for various geometries on $^{90}$Y [Coursey 1993] and $^{117m}$Sn, $^{62}$Cu, $^{188}$Re, $^{125}$I, $^{99m}$Tc, $^{133}$Xe, $^{153}$Gd, $^{153}$Sm and $^{18}$F [Zimmerman et al., 1998, 1999, 2000, 2001]. It was found that most of the dial settings recommended by the manufacturer result in an overestimation of the activity by up to 50%. In the case of $^{18}$F, up to 6% of overestimation of activity was found, depending on the sample geometries.

With the advent of pure β-particle emitters for therapeutic uses (e.g. $^{32}$P, $^{89}$Sr, $^{90}$Y), it is inevitable that the ionisation chambers of the radiopharmaceutical suppliers and users need to be standardised for these pure β-particle emitters. The ionisation chamber is not sensitive to β-particles. The measurement by ionisation chambers relies on measuring bremsstrahlung photons produced by the slowing down of the beta particles in the source, vial and chamber materials. Since the production of Bremsstrahlung and the corresponding energy distribution are highly dependent on the composition and amount of stopping material, measurement are likely to be affected by such factors as container type (vial, ampoule, syringe), and possibly solution density and container filling volume.
3.3. Neutron activated activity measurements in neutron flux determination and neutron activation analysis

Activity measurements are used in the nuclear science and engineering for the determination of neutron flux and cross section, and amounts of chemical elements in a sample. The later application is called Neutron Activation Analysis (NAA) which is based on the conversion of stable nuclei to other radioactive nuclei via neutron irradiations. It uses γ-ray spectrometry to identify and quantify the radioactive products. Since the cross sections of stable nucleus are accurately known, the amount of a specific element can be determined from the activity of specific products.

Neutron flux measurement is based on the one of the most important absorption reactions: radiative capture, or (n, γ) reaction. $^{197}$Au is commonly used as a neutron detector since [Hargrove and Geiger, 1964] 1) it is a monoisotopic element and can be obtained in high purity; 2) the decay scheme of $^{198}$Au is relatively simply and allows the absolute activity to be determined by coincidence counting; 3) the deviation from a perfect detector below cadmium cut-off energy is small; 4) the half-life of $^{198}$Au, equal to 2.6944 days, is short enough to allow reasonable irradiation times.

When exposed in a neutron field, $^{197}$Au captures a neutron and produces $^{198}$Au, followed by the emission of gamma radiation through the decay of $^{198}$Au into $^{198}$Hg. This process can be expressed by neutron capture reaction $^{197}$Au (n, γ) $^{198}$Au and β−γ decay $^{198}$Au → $^{198}$Hg + β− + γ. The neutron flux is determined through the reaction rate per gold atom, which is related to the activity by

$$R = \frac{MA(t)}{N_A m (1 - e^{-\lambda t})},$$

where, $M$ is the Molar mass of $^{197}$Au, $\lambda$ is the decay constant of $^{198}$Au, $N_A$ is the Avogadro number and $m$ is the mass of the gold detector.

Neutron flux measurements by gold activation have been performed during the “hot” commissioning for the ANSTO OPAL nuclear reactor.

4. Monte Carlo simulation technique in the activity measurements

The Monte Carlo simulation technique has overcome the shortcoming of the numerical method used previously for simple geometries based on the radiation transport equations. Although Monte Carlo simulation has been used for a few decades in many areas of experimental physics, such as elementary particles physics and high energy physics, it has only gained interest in radiation physics in recent decades due to the great advance of computing
power. This has allowed more complex physical processes to be simulated in a reasonable time. The availability of various codes has allowed Monte Carlo to be used extensively in: the investigation of detector response features; efficiency calibrations for different geometries; validating new designs; and improving the performance of new detectors for different applications.

Monte Carlo simulation in radiation physics simulates the transport of electrons, positrons and photons in materials with arbitrary compositions. It simulates the physical process of energy loss along the path of radiation particles. The “history” of each primary particle and of the secondary particle resulting from interaction of primary particle with the detector and surrounding materials is tracked until all its initial energy is dissipated (or below the cut-off energy). A large number of histories ($> 10^5$ to $10^6$) must be tracked to obtain satisfactory statistics, as only a small percentage of events leads to absorption.

The widely used codes include GEANT4 developed by CERN [Giani et al., 1998.], MCNP developed by the Los Alamos National Laboratory [Briesmeister, 1993], PENELOPE by the University of Barcelona [Salvat et al, 2001] and EGS4 by the Stanford Linear Accelerator Centre [Nelson et al, 1985]. The codes are more or less complicated, depending on the degree of sophistication in the geometry description and interactions modes taken into account. They generally include computing acceleration techniques and variance reduction methods. The Monte Carlo simulation may end up having more than 10% deviation from the experimental data. In this case, some detector parameters in the Monte Carlo simulation are adjusted to obtain better agreement [Kamboy and Kahn, 1996, Korun and Vidmar, 1997].

The codes have been used to calculate the mean energy deposited in the gas within the active volume of the $4\pi$ counter per photon emitted from the volume source. The Monte-Carlo code PENELOPE has been used to calculate ionisation chamber calibration factors (for $^{125}$I, $^{109}$Cd, $^{133}$Ba, $^{210}$Pb, $^{57}$Co, $^{99m}$Tc, $^{138}$Ce, $^{51}$Cr, $^{85}$Sr, $^{95}$Nb, $^{58}$Co, $^{54}$Mn, $^{65}$Zn, $^{59}$Fe, $^{56}$Co, $^{60}$Co, $^{22}$Na, and $^{88}$Y) and response function [De Vismes and Amiot, 2003]. The agreement between calculated and measured factors is better than 1%. The calculation of the calibration factors of radiopharmaceuticals with short half-lives, $^{18}$F, $^{99m}$Tc, $^{111}$In and $^{123}$I, has also been performed in the same laboratory [Amiot, 2004]. The response of $4\pi$ ionisation chamber has been simulated using MCNP code [Khalid, 2002], GEANT4 [Gostely and Leadermann, 2000] and EGS4 [Suzuki et al., 1998]. The Monte-Carlo code GEANT4 has been used to calculate the response curve vs photon energy and calibration factors for $^{60}$Co, $^{22}$Na and $^{59}$Fe for the ANSTO secondary standard ionisation chamber [Seneviratne, 2005]. Agreement within 0.2% of the experimental values for these nuclides has been achieved, except for $^{22}$Na, in which case annihilated photons need to be more carefully modelled in order to obtain higher degree of accuracy.
The Monte Carlo simulation is also used extensively to calculate the photopeak efficiency for HPGe and NaI detectors [Johnston and Burns, 1994, Hardy et al., 2002, Helmer et al., 2003, Salgado et al., 2006, Vlastou et al., 2006]. In the gamma spectrometry, the measured full energy photopeak efficiency must be corrected for cascade summing which occurs whenever two or more gamma rays from the same decay event are detected simultaneously inside the detector crystal. The cascade summing corrections have been performed using the Monte Carlo method [Dians et al, 2002].

The application of Monte Carlo simulation to the $4\pi\beta-\gamma$ coincidence counting was firstly carried out by Miyahara et al. [Miyahara et al, 1985]. In their work, the $\beta$ energy spectra and self-absorptions of $^{134}$Cs, $^{59}$Fe, $^{103}$Ru and $^{60}$Co spherical particle sources were calculated by using the Monte Carlo method. The Monte Carlo simulation of all detection processes for $^{134}$Cs in a $4\pi\beta-\gamma$ coincidence counting system was performed recently by Takeda et al. [Takeda et al, 2005]. Both of the above simulations allowed the calculation of the efficiency extrapolation function.

In liquid scintillation counting, Monte Carlo is used to calculate the probability for the interaction of X- or $\gamma$-photons in a given scintillator [Simpson, 1994] and the absorbed energy distribution [Cassette et al, 2004]. The absorbed energy distribution of photons is most often calculated in discrete energy bins and the quenching function applied to each bin midpoint [Zimmerman, 2006]. An overall efficiency for the X- or $\gamma$-photons is obtained by a summation over the energy spectrum. PENELOPE and EGSnrcMP package [Kawrakow, 2000, Kawrakow et al., 2003] (the new EGS4 version) have been used for the calculation.

Monte Carlo method for uncertainty evaluation

The interesting application of Monte Carlo method is to evaluate uncertainties of activities determined by TDCR method [Cassette et al, 2004] when a large number of transfer functions and parameters are involved. The principle of this method is to obtain the average and standard deviation from a large number of data sets generated from transfer functions through a large number of input data sets. The input data sets of the transfer function are characterised with their average value, standard deviation and supposed statistical distribution function. The authors computed 1000 data sets and 1000 activities using Gaussian distributions. The uncertainty for the detection efficiency of double coincidence was found to be 0.3% in this way.
5. Work associated with this thesis

The aim of this study was to standardise pure \( \beta \) and \( \beta-\gamma \) emitting radionuclides in various physical forms. The standardisation methods used in this work covered liquid scintillation TDCR and CIEMAT/NIST methods, \( 4\pi\beta-\gamma \) coincidence method, ionisation chamber measurement and gamma spectrometry. The techniques developed to deal with solid sources included destructive and non-destructive methods. Quantitative chemical digestion of the solid source was used in the destructive method, and Monte Carlo simulation was used as a mean of obtaining corrections for the non-destruction method.

The activity standards produced using the above techniques were used for the calibration of the ionisation chambers of the ANSTO Radiopharmaceutical Industries (ARI) and determination of neutron flux distributions for the commissioning of the ANSTO OPAL reactor.


This chapter describes the statistical and physical model used in the TDCR and CIEMAT/NIST methods and the theoretical and experimental approach for establishing detection efficiencies of a liquid scintillation system. A newly built ANSTO TDCR system and its performance test using \( ^3 \)H and \( ^{14} \)C sources representing low and high energies of beta particles are also demonstrated in this chapter.


This chapter describes the fundamental study of the detection efficiency model for the TDCR and CIEMAT/NIST methods.

According to the statistical and physical model of liquid scintillation counting, the discrimination level of the liquid scintillation system has to be set just before the single photoelectron peak so that all single photoelectron pulses caused by disintegration are counted.

However, this requirement cannot be met in some circumstances. For example, in the case of commercial liquid scintillation counters such as Wallac, Beckman and Parkard Tri-Carb, the discrimination level is set by the manufacturers and cannot be monitored or adjusted by users. If the level is incorrect due to any reason, it is not known how the activity determination would be influenced. Even for the TDCR system, when the peak-valley ratio is not high enough, it is very
likely that the discrimination level would be set too high and consequently some of the single photoelectron peak rejected.

The influence of rejection of a fraction of the single photoelectron peak has been studied both theoretically and experimentally on pure β-emitters \(^{3}\)H, \(^{63}\)Ni, \(^{14}\)C and \(^{90}\)Sr/\(^{90}\)Y. A modified formula for calculation of efficiency is given in the paper in consideration of the rejected fraction of the single photoelectron peak.

*Chapter Four: Development of activity standard for \(^{90}\)Y microspheres (published in Applied Radiation and Isotopes, vol 63, 2005, p193-199)*

For accurately measuring the activity of a solid source, a highly-quantitative destructive standardisation procedure is often used. The procedure involves dissolution of the solid source, quantitative solution transfers, and a gravimetric dilution, followed by using one or more of the above mentioned activity measurement techniques. Any activity loss during the process should be quantified. The destructive standardisation has to be performed by a unique radiochemical digestion procedure that was specifically designed for the source type.

This chapter describes how the activity standard of \(^{90}\)Y microspheres was developed by using the liquid scintillation TDCR and the CIEMAT/NIST methods in combination with a highly quantitatively destructive procedure.

*Chapter Five: Absolute activity determination of \(^{198}\)Au solid source using 4πβ–γ coincidence counting corrected by Monte-Carlo calculation (published in IEEE transactions on Nuclear Science, vol 54, No 3, June 2007)*

On some occasions, we cannot use a destructive method for activity measurements when a defined geometry has to be maintained to calibrate a secondary standard instrument. There is a need to explore a technique for a direct measurement of a solid source.

This chapter describes the absolute activity determination of \(^{198}\)Au in the form of Au wire, Al-Au (0.112% of Au) alloy wire and Au foil using the 4πβ–γ coincidence-counting method. A correction to the coincidence equation derived for the \(^{198}\)Au was determined using the Monte Carlo simulation technique. The Monte Carlo simulation was employed to simulate the complicated absorption and attenuation processes of electrons and gamma photon interactions with the surrounding materials. Probabilities of escape beta particles, internal conversion electrons and photon-interaction generated photoelectrons and Compton electrons calculated were used to determine the correction term of the coincidence equation.

This chapter describes the use of the activity standards, developed in the previous chapter, in the neutron flux measurements for the hot commissioning process of the ANSTO OPAL reactor. The activity standards were used to calibrate HPGe detectors for the 411 keV full energy photopeak. The thermal and epithermal neutron flux in the reactor core and irradiation facilities was determined through the activity measurements of activated Au monitors. The fast neutron flux in the irradiation facilities was determined through the activity measurements of activated Ni monitors.

Chapter Seven: Uncertainty evaluation of thermal neutron flux determination over the energy range 0 – 0.6 eV (published in the Proceeding of International Metrology Conference, April 3-7, 2006, Casablanca, Morocco)

Many fundamental and engineering studies in nuclear science depend on reliable radioactivity value. The accuracy in the determination of neutron flux, for example, is usually limited by uncertainties in the disintegration rate. This can be seen in the study, undertaken in this chapter, of the uncertainty analysis of the neutron flux described in the previous paper.


The positron emitter $^{18}$F is one of the most important imaging radionuclides in diagnostic nuclear medicine. The Australian National Medical Cyclotron (NMC) routinely produces ($^{18}$F)FDG. This chapter describes the calibration of the dose calibrator used by the NMC PET Quality Control (QC) Section for the $^{18}$F against the Australian primary standard. Calibration of this instrument is not as straightforward as the system which provides current readings from the electrometer. The “calibration curve method” was employed for the experimental determination of the dial setting number.

Chapter Nine: Development of an Australian secondary standard for the reference air kerma rate measurement of $^{125}$I seeds (published in Australasian Physical & Engineering Sciences in Medicine, vol 28, Nov. 3, 2005)
This chapter describes the establishment of the Australian secondary dosimetry standard for air kerma rate measurement of $^{125}$I brachytherapy seeds. A High-Dose-Rate 1000 Plus ionisation chamber was calibrated against the primary standard for an Amersham model 6711 $^{125}$I seed calibrated by the National Institute of Standards and Technology (NIST) in the USA.

Chapter Ten: Comparison of LaBr$_3$:Ce, LaCl$_3$:Ce, CZT and NaI(Tl) for Resolution of Nuclear Material Spectra (accepted by the IEEE-9th International Conference on Inorganic Scintillators and their Applications, June 4-8, 2007, Winston-Salem, USA)

New scintillators such as LaBr$_3$:Ce and LaCl$_3$:Ce, activated with cerium (Ce$^{3+}$) atoms as the primary scintillation source, have gained interest due to good resolution, high efficiency and fast response with high light output per keV. The scintillators have potential application in a $4\pi\beta-\gamma$ coincidence counting system for detection of photons.

This chapter describes the comparisons made of key parameters, such as energy resolution and detection efficiency, of LaBr$_3$:Ce and LaCl$_3$:Ce with NaI(Tl) and CdTeZn detectors.

Chapter Eleven: Activity measurements of the radionuclide $^{153}$Sm for the ANSTO, Australia in the ongoing comparison BIPM.RI(II)-K1.Sm-153 (published in Metrologia, vol 42, Technical Supplement, 06008)

The activity standard of $^{153}$Sm developed at the ANSTO was compared with those developed by other countries through the International Reference System (SIR) managed at the BIPM (Bureau International des Poids et Mesures) in the France. The results show that the ANSTO standard agrees with the Key Comparison Reference Value (KCRV) within the combined standard uncertainties.

References


Wyllie, H.A., 1989. The preparation of radioactive sources with radioactivities of less than 110 Kilobecquerels, ANSTO/M118.


Chapter Two

Development of new capabilities in radionuclide measurements using TDCR method within ANSTO

L, Mo, J.M.Qin, M. Hurry

Australian Nuclear Science and Technology Organisation

Author’s contribution

L. Mo (candidate)
Investigated the methodology, planed and led the project, sourced the best design of TDCR system in the world, wrote the manuscript.

J.M. Qin
Built the ANSTO TDCR system and drew the diagrams.

M. Hurry
Built the ANSTO TDCR system and drew the diagrams.
Development of new capabilities in radionuclide measurements using TDCR method within ANSTO

L. Mo
M. J. Qin
Mark Hurry

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ABSTRACT

The TDCR (Triple to Double Coincidence Ratio) method for the activity measurement of pure beta emitting nuclides and the newly constructed ANSTO TDCR system, as well as its performance test using $^3$H and $^{14}$C, are presented in this paper. Measured activities for $^3$H and $^{14}$C, representing low and high energy pure beta emitters, agree with the activity value obtained at the Laboratoire National Henri Becquerel (LNHB) to better than 0.5% and 0.3% respectively. The results indicate that the ANSTO TDCR system is capable of providing accurate activity measurements for pure beta emitting nuclides.
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1. Introduction

The triple to double coincidence ratio (TDCR) and CIEMAT/NIST method, named after the two institutes who originate it: Centro de Investigaciones Energeticas Medioambientales y Tecnologicas in Spain and National Institute of Standards and Technology in US, is widely used in the liquid scintillation (LS) counting to standardise a pure $\beta$-emitting nuclide. The TDCR method is a direct method because of that it does not require a reference standard, although it does rely on decay scheme parameters to calculate efficiencies. The method utilizes a three photomultiplier tubes (PMTs) system in which the three PMTs are mounted at 120° intervals around the scintillation vial in the same horizontal plane. The CIEMAT/NIST method utilizes a commercial two PMT system in which the two PMTs are mounted at 180° intervals. The CIEMAT/NIST method is a relative method which uses a tritium quenching standard to determine a quench curve for the nuclide of interest. Although not a direct activity measurement method, it allows relative standard uncertainties of less than 1% to be achieved even for low-energy $\beta$-emitters ($E_{\beta_{\text{max}}} > 50$ keV).

The first three-PMT system was constructed by Hoegl and Schwerdtel [Hoegl and Schwerdtel, 1963] in 1963. However this system was used for relative measurements of pure $\beta$-emitters [Schwerdtel, 1966a, b]. In 1970, Kolarov et al. [Kolarov et al., 1970] suggested an absolute method of determining the activity of $\beta$-emitters based on a simple detection efficiency model using two phototubes working in sum and in coincidence. The major disadvantages of this method were the effect of afterpulses and thermal emission noise observed in the sum channel.

The above triple coincidence LS detector and the principle of the detection efficiency model have both been the basis of the TDCR absolute method of radionuclide activity determination.

The original version of TDCR method was presented by Pochwalski and Radoszewski (Pochwalski and Radoszewski, 1979, Pochwalski, et al. 1988) in 1979. It was based upon linear extrapolation of the experimental triple to double coincidence ratio (TDCR) to unity. The TDCR was varied by change of the optical efficiency.

The recently developed TDCR method [Vatin, 1991, Cassette and Vatin, 1992, Broda and Pochwalski, 1992, Meyer and Simpson, 1990] equalises the measured TDCR with the theoretical expressions to obtain the detection efficiency. This method does not need extrapolation for determining activity. However, it still requires efficiency variation for the best estimation of the ionisation quenching parameter $k_B$ value.

In the early 1990s, the electronics of the TDCR system was modified to introduce three additional double coincidence counting outputs $N_{ab}$, $N_{bc}$ and $N_{ac}$ [Vatin, 1991, Cassette and
Vatin, 1992, Broda and Pochwalski, 1992] in addition to the two described previously and in reference [Pochwalski, et al. 1988]. This allows three more equations to be established and thus the assumption of symmetrical behaviour of the three phototubes is no longer necessary. In fact, the real asymmetry can only be ignored for high counting efficiency. Errors introduced by the symmetry simplification increase when the efficiency decrease [Grau Malonda and Coursey, 1988]

Broda [Broda, 2003] has given an excellent review on the development of TDCR method.

The CIEMAT/NIST method originated in 1982, when Grau Malonda and Garcia-Torano [Grau Malonda and Garcia-Torano, 1982] presented a method for the calculation of counting efficiency for $\beta$-ray emitting nuclides in two-phototube system. The author used a set of tritium standards to obtain an experimental quenching curve, from which the $\beta$-particle counting efficiency was calculated. Tritium is a low-energy $\beta$-ray emitter. Due to the low energy, the uncertain in tritium standard has little influence to the calculated efficiency of the more energetic $\beta$-ray emitters when using the tritium quenching parameter. In the same year, the author extended this method to electron capture nuclides which was more complicated than $\beta$-ray emitters [Grau Malonda, 1982]. Different ways of atomic rearrangement and their probabilities have to be considered. A Monte-Carlo code had to be used for computing the escape probability of an X-ray.

In 1986, Coursey et al. [Coursey, et al., 1986] from NIST and CIEMAT elaborated this method with more detailed processes and studies. Since then, it has been widely used for standardizing $\beta$-ray emitting radionuclide.

2. ANSTO TDCR system

TDCR method is applied to a three-photomultiplier tube system for liquid scintillation counting. Such a system is not available commercially. CIEMAT/NIST method is applied to a commercial two-photomultiplier tube system.

ANSTO has recently completed the construction of a TDCR system. The system was constructed on the basis of the NIST (the National Institute of Standards and Technology), LNHB (Laboratoire National Henri Becquerel) and BIPM (Bureau International Des Poids et Mesures) TDCR system. Figure 1 shows the block diagram of the ANSTO TDCR system. The system consists of an optical chamber, three photomultiplier tubes and signal process electronic units. The optical chamber is made of aluminium. The inside surfaces of the chamber are painted with reflective paint (titanium dioxide) to increase the light collection efficiency. Three 8850 Burle photomultiplier tubes are mounted at 120° intervals around the liquid scintillation
vial. The distance between the photomultiplier tube and vial is approximately 1 cm. The voltage
dividers are constructed in-house. Figure 2. shows the electronic circuit scheme of the voltage
dividers.

Signals from the anode of a PMT are used for counting. The output of the anode is
connected to a CAEN Model N412 8-channel Fast Amplifier. The MAC-3 (module
d’acquisition de coincidences triples [Bouchard and Cassette, 2000]) unit, supplied by the
LNHB, is used to process all the signals from the amplifier. MAC-3 is an electronic coincidence
module which replaces the complex interconnection of nine NIM independent electronic
instruments and gives five outputs (TTL singles): three double coincidences, AB, BC and CA,
one triple coincidence ABC and a logic sum of the three double coincidences including triple
coincidences, AB+BC+CA. The module uses extendable deadtime circuitry so that system live
time during counting period can be obtained. Therefore, no deadtime correction is required. The
extendable deadtime is at least 40 μs. The counting data are acquired using two National
Instruments PCI6602 Counter/Timers.

Signals from D9 of the PMT are amplified and fed to a Canberra ADC 8715 for acquiring
liquid scintillation spectrums. The single event outputs A, B, C of the MAC-3 are connected to
the GATE input of the Canberra ADC 8715 one at a time. By observing the spectra while
varying the discriminators in the MAC-3, the discrimination level of MAC-3 for each PMT
output channel is adjusted. The discrimination level of the system should be set just before the
single photoelectron peak so that all single photoelectron pulses caused by disintegration are
counted. The discrimination levels for three PMT outputs are set sequentially.

A LabView program has been written for data acquisition, high voltage supply to the anode
and focusing electrode of each PMT.

3. Ionisation quenching

If an ionising particle deposits energy E in a very short track length x (high dE/dx), there is
a reduction in the scintillation efficiency. This loss was described by Birks [Birks 1958] as
“ionisation quenching”. Birks formulated a semi-empirical model for the rate of light output dL
per unit track length dx (number of emitted photons per unit distance along the path) as
following,

\[
\frac{dL}{dx} = \frac{\eta_I dE/dx}{1 + k_B (dE/dx)}
\]

(1)
where, $\eta_s$ is the scintillation efficiency (number of fluorescence photons emitted per unit of energy), $k$ is the rate constant for ionisation quenching, $dE/dx$ is the stopping power (the specific energy loss, normally expressed in MeV cm$^{-2}$ g$^{-1}$), and $BdE/dx$ is the concentration of ionising events (or the linear ionization density).

The specific fluorescence in the absence of ionization quenching is:

$$\frac{dL}{dx} = \eta_s \frac{dE}{dx} \quad (2)$$

Therefore, $(1 + kB \frac{dE}{dx})^{-1}$ is the reduction factor due to the ionisation quench and $kB$ is the ionization quench parameter.

The integration of equation (2) gives:

$$L = \eta_s \int_0^R \left(\frac{dE}{dx}\right) dx = \eta_s \int_0^E dE = \eta_s E . \quad (3)$$

Therefore $\eta_s = L/E$ is the figure of merit of the scintillator when $kB(dE/dx)$ is negligible (compared with 1). For electrons with energy higher than 1 MeV, $dE/dx \rightarrow$ zero.

When $kB(dE/dx)$ is not negligible, we have

$$L(E) = \eta_s \int_0^E \frac{dE}{1 + kB(dE/dx)} = \eta_s E Q(E) , \quad (4)$$

where

$$Q(E) = \frac{1}{E} \int_0^E \frac{dE}{1 + kB(dE/dx)} \quad (5)$$

is called ionisation quench function or ionisation quench correction factor. It can be obtained by numerical integration of equation (5). $kB$ value, the ionization quench parameter characterising the scintillator, is in the range of 0.005 and 0.015 cm·g/MeV.

Thus, the figure of merit will be in this case,

$$\eta(E) = \frac{L(E)}{E} = \eta_s Q(E) . \quad (6)$$

From this equation, one can see that the figure of merit is consisted of two factors: $\eta_s$, independent of the energy and $Q(E)$, a function of the energy but dimensionless.

The formulae of the linear collision stopping power for electron, $S_{col}$ (MeV cm$^{-1}$), is given by (ICRU Report, 1984)
\[
\frac{dE}{dx} = \frac{2m_e^2 mc^2 \rho}{\mu} \frac{1}{\beta^2} \frac{Z^2}{A} \ln(T/I)^2 + \ln(1 + \tau / 2 + F^{-}(\tau) - \delta), \tag{7}
\]

where
\[
F^{-}(\tau) = (1 - \beta^2)\left[\frac{1}{2} + \frac{\tau^2}{8} - \frac{(2\tau + 1)\ln 2}{2}\right]. \tag{8}
\]

Where
- \(r_e\): classical electron radius (cm),
- \(mc^2\): electron rest energy (MeV),
- \(\rho\): density of the medium (g cm\(^{-3}\)),
- \(\mu\): atomic mass unit, \(1.6605055 \times 10^{-24}\) g (1/12 of the mass of an atom of the nuclide \(^{12}\)C),
- \(\beta\): velocity of the incident particle (projectile) divided by the velocity of light,
- \(Z\): atomic number,
- \(A\): relative atomic mass,
- \(I\): mean excitation energy of the medium,
- \(T\): kinetic energy of the incident electron,
- \(\tau\) = \(T/mc^2\), the ratio of the kinetic energy of the incident electron to its rest energy.
- \(\delta\): density-effect correction.

Equation (4) gives the mean number of photos produced in the scintillator when an electron deposits its energy \(E\) in the liquid scintillator. It demonstrates that the dependence of the number of photons of individual scintillations upon incident electron energy \(E\) is not linear. At low energy the deviation from linearity is caused by ionisation quenching which occurs when the specific energy loss (dE/dx) is large and according to Ref. [Gibson and Gale 1968], it can be described by the function \(Q(E)\).

4. Detection efficiency calculations

Both TDCR and CIEMAT/NIST methods for the calculation of the detection efficiency are based on the same statistical and physical models of the counting process and three assumptions [Cassette, 2002]: 1) the distribution of photons emitted by a scintillator when absorbing a monoenergetic electron follows Poisson distribution; 2) the detector threshold is lower than the amplitude of the single photoelectron peak, so the detection probability of any single photon is not zero, and 3) the non-linearity of the scintillation efficiency due to ionization-quenching can be described by Birks law [Birks, 1958].
Based on the assumption 1), for the electron energy $E$ deposited in the liquid scintillator, the probability of emission of $x$ photons with the mean number of photons being $m$ is given by

$$P(x / m) = \frac{m^x e^{-m}}{x!}$$

(9)

The non-detection probability is the probability of obtaining 0 photons for the mean of $m$. The detection efficiency is the complement of the non-detection probability and can be expressed by

$$\text{detection efficiency} = 1 - P(0 / m) = 1 - e^{-m}$$

(10)

Equation (4) gives the mean number of photos produced in the scintillator when an electron deposits its energy $E$ in the liquid scintillator, according to assumption 3),

$$m(E) = \int_0^\infty \frac{\eta_s dE}{1 + kB(\frac{dE}{dx})} = \eta_s E Q(E)$$

(11)

The distribution of photoelectrons obtained at the photocathode of a phototube is a result of three random processes: Poisson distribution for the light emission, multinomial distribution for the behaviour of photons inside the optical chamber and binomial distribution for the photoelectric process at the photocathode. Applying a quantum efficiency (overall optical efficiency) $\nu$, the distribution of photoelectrons at the photocathode can be described by a Poisson distribution and the detection efficiency becomes

$$\text{detection efficiency} = 1 - P(0 / m) = 1 - e^{-\nu m}$$

(12)

Let $\lambda = \nu \eta_s$ be the figure of merit (number of photoelectrons emitted at photocathode per unit of energy deposited in the scintillator), equation (12) becomes

$$\text{detection efficiency} = 1 - P(0 / m) = 1 - e^{-\lambda E Q(E)}$$

(13)

For a two-photomultiplier tube system, the detection efficiency of the two photomultiplier tubes with the same quantum efficiency $\nu$ in coincidence is given by

$$\text{detection efficiency} = \left[1 - e^{-\lambda E Q(E)}\right]^2$$

(14)

The above formula shows the calculated detection efficiency for the coincidence events registered in two phototubes for the mean number of photoelectrons produced at photocathode by mono-energetic electron in the scintillator. An electron energy spectrum generated by a radionuclide can be described by a normalised density function $S(E)$. The detection efficiency in coincidence is thus given by
Detection efficiency = \( \int_{E_0}^{E_{\text{max}}} S(E) \left[ 1 - e^{-\frac{\lambda E}{E}} \right]^2 dE \)  \( (15) \)

For a three-photomultiplier tube system, the detection efficiency of logical sum of double coincidence is given by

\[ \varepsilon_D = \int_{E_0}^{E_{\text{max}}} S(E) \left[ 3(1 - e^{-\frac{\lambda E}{E}})^2 - 2(1 - e^{-\frac{\lambda E}{E}})^3 \right] dE \]  \( (16) \)

and of triple coincidence is given by

\[ \varepsilon_T = \int_{E_0}^{E_{\text{max}}} S(E) \left[ 1 - e^{-\frac{\lambda E}{E}} \right]^3 dE \]  \( (17) \)

and therefore TDCR is given by

\[ \text{TDCR} = \frac{\varepsilon_T}{\varepsilon_D} = \frac{N_T}{N_D} = \frac{\int_{E_0}^{E_{\text{max}}} S(E) \left[ 1 - e^{-\frac{\lambda E}{E}} \right]^3 dE}{\int_{E_0}^{E_{\text{max}}} S(E) \left[ 3(1 - e^{-\frac{\lambda E}{E}})^2 - 2(1 - e^{-\frac{\lambda E}{E}})^3 \right] dE} \]  \( (18) \)

The detection efficiency for both of CIEMAT/NIST and TDCR method can therefore be calculated once the figure of merit is determined. The figure of merit is independent of the energy and is a function of the chemical quenching state for a particular system. The CIEMAT/NIST method uses a \(^3\)H quenching standard to determine the figure of merit when a two photomultiplier tube system is used. While with the TDCR method, the TDCR value in equation (19) is determined experimentally, and is equal to the ratio of the triple to logical sum of the double coincidence counting rate. Solving equation (18) then gives the value of the figure of merit.

For pure beta emitters, \( S(E) \) can be calculated by the Fermi function \([\text{Evans, 1955, Wu, 1966, Behrens and Szybisz 1976}]\).

From equation 1 it can be seen that \( k_B \) is the coefficient of the linear energy transfer (LET) \( dE/dx \). A precise evaluation of the \( k_B \) value is difficult but a best estimate can be obtained by a self-consistent calculation procedure for various values of the detection efficiency. Plotting activity values calculated from different efficiencies vs experimental TDCR, the best \( k_B \) value is obtained from the one that gives constant activity value over a range of TDCR. In another word the activity is independent of the TDCR.

There are several methods of varying the detection efficiency: chemical quenching, use of coaxial grey filters and. The first two processes reduce the mean quantity of light emission. The
last reduces the detection probability. Cassette et al. [Cassette et al, 2000] indicated that the three procedures were equivalent. However, defocusing is more practical over other two processes.

Detection efficiency variation can be achieved using different techniques: chemical quenching, use of coaxial grey filters and defocusing of photomultiplier tubes by reducing the focusing electrode potential. The first two processes reduce the mean quantity of light emission. The last reduces the detection probability. Cassette et al. [Cassette et al, 2000] indicated that the three procedures were equivalent. However, defocusing is more practical over other two processes. There is a limited range in which the focusing electrode potential can be changed. For a 2700 V potential at the anode with the photocathode being grounded, the optimum first dynode and focusing electrode potentials are around 800 V. The focusing electrode potential can be decreased to 600 V. Below it, the measurement data are not good at all.

5. Asymmetric model

Three phototubes with the same quantum efficiency are very difficult to get. One often obtains three very different single (A, B and C) and double count rates (AB, BC and CA). For three phototubes A, B and C with quantum efficiencies of $\nu_1$, $\nu_2$ and $\nu_3$, or figure of merit $\lambda_1$, $\lambda_2$ and $\lambda_3$, the double efficiency for each pair of phototubes are

$$
\varepsilon_{AB} = \int_0^{E_{\text{max}}} S(E) \left[ 1 - e^{-\lambda_B E} \right] \left[ 1 - e^{-\lambda_C E} \right] dE
$$

$$
\varepsilon_{BC} = \int_0^{E_{\text{max}}} S(E) \left[ 1 - e^{-\lambda_A E} \right] \left[ 1 - e^{-\lambda_C E} \right] dE
$$

$$
\varepsilon_{AC} = \int_0^{E_{\text{max}}} S(E) \left[ 1 - e^{-\lambda_A E} \right] \left[ 1 - e^{-\lambda_B E} \right] dE
$$

the logical sum of double coincidence efficiency is

$$
\varepsilon_D = \int_0^{E_{\text{max}}} S(E) \left[ \left( 1 - e^{-\lambda_A E} \right) \left( 1 - e^{-\lambda_B E} \right) + \left( 1 - e^{-\lambda_A E} \right) \left( 1 - e^{-\lambda_C E} \right) + \left( 1 - e^{-\lambda_B E} \right) \left( 1 - e^{-\lambda_C E} \right) \right] dE
$$

and of triple coincidence is given by

$$
\varepsilon_T = \int_0^{E_{\text{max}}} S(E) \left[ 1 - e^{-\lambda_A E} \right] \left[ 1 - e^{-\lambda_B E} \right] \left[ 1 - e^{-\lambda_C E} \right] dE.
$$

From equation (19), (20), (21) and (23), we can obtain the ratio of triple to double coincidence $\frac{\varepsilon_T}{\varepsilon_{AB}}$, $\frac{\varepsilon_T}{\varepsilon_{BC}}$ and $\frac{\varepsilon_T}{\varepsilon_{AC}}$. 
Similarly, the above calculated ratios of triple to double coincidence efficiencies are let to be equal to the experimental ratio of triple to double coincidence counting rate. The free parameters for each phototube are found by solving these three equations and then used to calculate the detection efficiency for the logical sum of the double coincidence, from which the source activity is determined.

An electronic module MAC3 (module d’acquisition de coincidences triples) has been developed to process the pulses delivered by the three phototubes [Bouchard and Cassette, 2000] giving five output: three double coincidence, AB, BC and CA, a triple coincidence ABC (T) and a logical sum of the three double coincidence AB + BC + CA. This module, replaces the complex interconnection of nine NIM independent and costly electronic instruments. Four count rates, \(N_{AB}\), \(N_{BC}\), \(N_{CA}\) and \(N_T\), are sufficient to calculate the disintegration rate. The fifth, \(N_D\), is used exclusively for the test of counting correctness [Broda, 1992], which is

\[
N_{AB} + N_{BC} + N_{CA} = 2N_T + N_D.
\]

6. Validation of ANSTO TDCR system

\(^3\)H contained in an ampoule was supplied by the LNHB with a standardisation certificate. 38 to 52 mg of radioactive mass was dispensed into ordinary commercial type of 20ml liquid scintillation vials to make up 10 ml radioactive solutions. The scintillator used was Ultima Gold cocktail. Five \(^3\)H samples were prepared. \(^{14}\)C source in a liquid scintillation vial was also supplied by LNHB with a standardisation certificate. No sub-dispensing of the \(^{14}\)C source was made. The scintillator for \(^{14}\)C was Ultima Gold cocktail.

The discrimination levels were set just below the single photoelectron peak to count all pulses. The detection efficiency was changed by varying the potential of the focusing electrode from 680, in increment of 20 V to 800 V. Each source was counted for 10 times of 60s duration for each setting.

Plotting activity values calculated from different efficiencies vs experimental TDCR, the best \(k_B\) value was found to be 0.012 for both \(^3\)H (Figure 3) and \(^{14}\)C. Actually, \(k_B\) value is insensitive to efficiencies for \(^{14}\)C due to high energy of beta particles. Using these \(k_B\) values the experimental TDCR value was found to be 0.40 and 0.91 for \(^3\)H and \(^{14}\)C respectively. The partial TDCR such as T/AB, T/BC and T/AC were 0.68, 0.68 and 0.64 respectively for \(^3\)H and 0.97, 0.97 and 0.97 for \(^{14}\)C.
Using the kB and TDCR values described above and efficiency equation (22), the activities of $^3$H and $^{14}$C were found to agree with the activity value obtained at the Laboratoire National Henri Becquerel (LNHB) to better than 0.5% and 0.3% respectively.

7. Summary

TDCR and CIEMAT/NIST methods are important methods in liquid scintillation counting for the determination of the activity of pure beta emitting and pure electron capture nuclides. Both of them are based on the same physical principle and statistical assumption of the photon distribution emitted in the scintillator and their detection probability in the photomultiplier tube. The determination of detection efficiency for both methods involves obtaining figure of merit for a particular experimental condition. TDCR method is applied to a three-photomultiplier tube system and is a direct method. Figure of merit is obtained by letting the experimental TDCR value equals to the theoretical TDCR. CIEMAT/NIST method is applied to a commercial two-photomultiplier tube system and is a relative method. Figure of merit is found by using a tritium reference standard. Both methods have a high accuracy in the determination of detection efficiency.

$^3$H and $^{14}$C measured in the newly built ANSTO TDCR system were found to agree with the activity value obtained at the Laboratoire National Henri Becquerel (LNHB) to better than 0.5%
and 0.3\% respectively. The results indicate that the ANSTO TDCR system is capable of providing accurate activity measurements for pure beta emitting nuclides.

**References:**


Pochwalski, K., Radoszewski, T., 1979. Disintegration rate determination by liquid scintillation counting using the triple to double coincidence ratio (TDCR) method. Institute of Nuclear Research, Warsaw, INR 1848/OPiDI/E/A.


Figure 1. Block diagram of the ANSTO TDCR system.
Figure 2. Block diagram of voltage divider.
Chapter Three

The influence of rejection of a fraction of the single photoelectron peak in liquid scintillation counting

L. Mo\textsuperscript{a, c}, P. Cassette\textsuperscript{b}, C. Baldock\textsuperscript{c}

\textsuperscript{a}Australian Nuclear Science and Technology Organisation, New Illawarra Road, Lucas Heights, NSW 2234, Australia

\textsuperscript{b}CEA/BNM-Laboratoire National Henri Becquerel, CEA-Saclay, Gif-sur-Yvette Cedex, F91191, France

\textsuperscript{c}Institute of Medical Physics, School of Physics, University of Sydney, NSW 2006, Australia

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Author’s contribution

L. Mo (candidate)
Designed and carried out experiment, analysed theoretical model and experimental data, and wrote manuscript.

P. Cassette
Designed experiment and modified theoretical model.

C. Baldock
Edited manuscript.
The influence of rejection of a fraction of the single photoelectron peak in liquid scintillation counting

L. Mo\textsuperscript{a,c,}, P. Cassette\textsuperscript{b}, C. Baldock\textsuperscript{c}

\textsuperscript{a}Australian Nuclear Science and Technology Organisation, New Illawarra Road, Lucas Heights, NSW 2234, Australia
\textsuperscript{b}CEA/BNM-Laboratoire National Henri Becquerel, CEA-Saclay, Gif-sur-Yvette Cedex, F91191, France
\textsuperscript{c}Institute of Medical Physics, School of Physics, University of Sydney, NSW 2006, Australia

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Abstract

Zero discrimination level setting of liquid scintillation systems for counting of all single photoelectron pulses is required by both triple-to-double coincidence ratio (TDCR) and CIEMAT/NIST methods. However, this requirement may not be able to be met in some circumstances. The influence of rejection of a fraction of the single photoelectron peak has been studied both theoretically and experimentally on \textsuperscript{3}H, \textsuperscript{63}Ni, \textsuperscript{14}C and \textsuperscript{90}Sr/\textsuperscript{90}Y for both the TDCR and CIEMAT/NIST methods. A modified formula for calculation of efficiency is given in consideration of the rejected fraction of the single photoelectron peak.

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Keywords: TDCR; CIEMAT/NIST; Single photoelectron peak; Fraction correction; \textsuperscript{3}H; \textsuperscript{63}Ni; \textsuperscript{14}C and \textsuperscript{90}Sr/\textsuperscript{90}Y

1. Introduction

It is well known that with either the triple-to-double coincidence ratio (TDCR) method [1–5] or CIEMAT/NIST method [6–8], the discrimination level has to be set just before the single photoelectron peak so that all single photoelectron pulses caused by disintegration are counted as required by the detection efficiency calculation model. If a significant fraction of these pulses is rejected by the discriminator, the experimental detection efficiency is no longer equal to the theoretically calculated detection efficiency, and an error may occur in the obtained parameter, the figure of merit.

Commercial liquid scintillation counters such as Wallac, Beckman and Parkard Tri-Carb employ the CIEMAT/NIST method to standardise radionuclides using a tritium quenching standard to determine the quench curve for the nuclide of interest. However unlike a TDCR system, the discrimination level for such counters is set by the manufacturers and cannot be monitored or adjusted by users. If the level is incorrect due to any reason, it is not known how the activity determination would be influenced.

Four beta emitters \textsuperscript{3}H, \textsuperscript{63}Ni, \textsuperscript{14}C and \textsuperscript{90}Sr/\textsuperscript{90}Y were chosen to study the influence of rejection of a fraction of the single photoelectron peak on the figure of merit and activity. The study of \textsuperscript{3}H is of particular interesting since it is a tracer radionuclide in the CIEMAT/NIST method. \textsuperscript{63}Ni is a low-energy beta emitter, \textsuperscript{14}C is a middle to high-energy beta emitter and \textsuperscript{90}Sr/\textsuperscript{90}Y are high-energy beta emitters.
2. Theoretical considerations

2.1. Modified formula for detection efficiency calculation

The calculation of the detection efficiency for both CIEMAT/NIST and TDCR methods is based on the same statistical and physical models of the counting process and three assumptions [10]: (1) the distribution of photons emitted by a scintillator when absorbing a monoenergetic electron follows Poisson distribution; (2) the detector threshold is lower than the amplitude of the single photoelectron peak, so the detection probability of any single photon is not zero; and (3) the non-linearity of the scintillation efficiency due to ionisation-quenching can be described by Birks law [11].

Here we extend assumption (2) to the situation where the discrimination level is set to reject a fraction of the single photoelectron peak, so the detection probability of some single photons becomes zero. In this situation the detection efficiency can still be accurately calculated if modification to the conventional formula is undertaken as described as below.

For the electron energy \( E \) deposited in the liquid scintillator, the probability of emission of \( x \) photons with the mean number of photons being \( m \) is given by

\[
P(x/m) = \frac{m^x e^{-m}}{x!}. \tag{1}
\]

The non-detection probability is the probability of obtaining 0 photons for the mean of \( m \). The detection efficiency is the complement of the non-detection probability and can be expressed by

\[
ev_D = \int_{\text{spectrum}} S(E) \left[ 1 - (1 + f \lambda EQ(E)/3)e^{-\lambda EQ(E)/3} \right]^3 \, dE.
\]

(3)

\[
\text{TDCR} = \int_{\text{spectrum}} S(E) \left[ 1 - (1 + f \lambda EQ(E)/3)e^{-\lambda EQ(E)/3} \right]^3 \, dE
\]

(4)

where \( A \) is the scintillation efficiency (number of fluorescence photons emitted per unit of energy), \( k \) is the rate constant for ionisation quenching, \( B dE/dx \) is the linear ionisation density, and

\[
Q(E) = \frac{1}{E} \int_0^E \frac{dE}{1 + kE(dE/dx)}
\]

(5)

is the ionisation quench function. Let \( \lambda = vA \) be the figure of merit (number of photoelectrons emitted at photocathode per unit energy deposited in the scintillator), Eq. (3) then becomes

\[
ev_D = \int_{\text{spectrum}} S(E) \left[ 1 - (1 + f \lambda EQ(E)/2)e^{-\lambda EQ(E)/2} \right]^2 \, dE.
\]

(6)

For a three-photomultiplier tube system, the detection efficiency of logical sum of double coincidence is given by

\[
ev_D = \int_{\text{spectrum}} S(E) \left[ 3 \left( 1 - (1 + f \lambda EQ(E)/3)e^{-\lambda EQ(E)/3} \right)^2 - 2 \left[ 1 - (1 + f \lambda EQ(E)/3)e^{-\lambda EQ(E)/3} \right]^3 \right] \, dE
\]

(7)

and of triple coincidence is given by

\[
ev_T = \int_{\text{spectrum}} S(E) \left[ 1 - (1 + f \lambda EQ(E)/3)e^{-\lambda EQ(E)/3} \right]^3 \, dE
\]

(8)

and therefore TDCR is given by

\[
\text{TDCR} = \frac{\int_{\text{spectrum}} S(E) \left[ 1 - (1 + f \lambda EQ(E)/3)e^{-\lambda EQ(E)/3} \right]^3 \, dE}{\int_{\text{spectrum}} S(E) \left[ 1 - (1 + f \lambda EQ(E)/3)e^{-\lambda EQ(E)/3} \right]^2 \, dE - 2 \left[ 1 - (1 + f \lambda EQ(E)/3)e^{-\lambda EQ(E)/3} \right]^3 \, dE}.
\]

(9)

The detection efficiency for both of CIEMAT/NIST and TDCR method can therefore be calculated once the figure of merit is determined. The figure of merit is independent of the energy and is a function of the chemical quenching state for a particular system. The CIEMAT/NIST method uses a \(^{3}\)H quenching standard to determine the figure of merit when a two photomultiplier tube system is used. While with the TDCR method, the TDCR value in Eq. (9) is determined experimentally, and is equal to the ratio of the triple to logical sum of the double coincidence counting rate. Solving Eq. (9) then gives the value of the figure of merit.

2.2. The discrepancy in figure of merit and activity when no correction is made

The detection efficiency for low-energy beta emitters varies more greatly with the figure of merit \( \lambda \) than high-
energy beta emitters in the range of $\lambda > 0.3$ e/keV where the figure of merit is usually within. The calculated TDCR value and detection efficiency of logical sum of double coincidence, for a three-photomultiplier tube system, as a function of the figure of merit is plotted in Fig. 1(a) for $^{3}$H and $^{90}$Sr/$^{90}$Y. Fraction correction is applied to the calculation for $f = 0.5\%$.

If no correction is made for the rejection of a fraction of the single photoelectron peak, the determined figure of merit will have a discrepancy. Fig. 2(a) shows the calculated discrepancies in figure of merit as a function of rejected fraction of the single photoelectron peak for TDCR method for $^{3}$H and $^{63}$Ni, and (b) $^{14}$C and $^{90}$Sr/$^{90}$Y. Discrepancies for a two-photomultiplier tube system for $^{3}$H as a quenching standard is also shown in (a). The true figure of merit is assumed to be 0.7932 e/keV.

One can calculate the discrepancy in the activity of a source due to rejection of a fraction of the single photoelectron peak if no correction to the efficiency calculation model is made. When a source with an activity of $A$ is counted in a three-photomultiplier tube system under the normal condition of zero discrimination, from the experimental and calculated TDCR value, the true figure of merit $\lambda$ is obtained from Eq. (9) with $f = 0$. When a fraction $f$ of the single photoelectron peak is rejected by increasing the discrimination level, the experimental triple to double coincidence ratio would be reduced to $R_f$. Since the figure of merit is the parameter characterising the liquid scintillator and photomultiplier tube, it should not change with the variation of the threshold of the pulse amplitude in the electronic pulse process system. Using fraction-corrected efficiency calculation formula (9), $R_f$ can be found for a given $\lambda$ and $f$. If $R_f$ is let to equal to the right hand side of Eq. (9) with $f = 0$ (no fraction correction),
that is

\[
R_f = \frac{\int_{\text{spectrum}} S(E) \left[ 1 - (1 + f \lambda E Q(E)/3) e^{-\lambda E Q(E)/3} \right]^3 \, dE}{\int_{\text{spectrum}} S(E) \left[ 1 - e^{-\lambda E Q(E)/3} \right]^3 \, dE}
\]

\[
= \frac{\int_{\text{spectrum}} S(E) \left( 3 \left[ 1 - (1 + f \lambda E Q(E)/3) e^{-\lambda E Q(E)/3} \right]^2 - 2 \left[ 1 - (1 + f \lambda E Q(E)/3) e^{-\lambda E Q(E)/3} \right] \right) \, dE}{\int_{\text{spectrum}} S(E) \left( 3 \left[ 1 - e^{-\lambda E Q(E)/3} \right]^2 - 2 \left[ 1 - e^{-\lambda E Q(E)/3} \right] \right) \, dE}
\]

(10)

the figure of merit \( \lambda_f \) obtained would be lower than the true value \( \lambda \) (\( \lambda_f < \lambda \)) along with the double efficiency. Consequently, the activity would have a discrepancy (in ratio) of

\[
A_1 = \frac{\varepsilon_D}{\varepsilon_T} = \frac{\int_{\text{spectrum}} S(E) \left[ (1 - (1 + f \lambda E Q(E)/3) e^{-\lambda E Q(E)/3}) \right]^3 \, dE}{\int_{\text{spectrum}} S(E) \left[ 1 - e^{-\lambda E Q(E)/3} \right]^3 \, dE}
\]

(12)

and

\[
A_1 = \frac{\varepsilon_d}{\varepsilon_T} = \frac{\int_{\text{spectrum}} S(E) \left[ (1 - (1 + f \lambda E Q(E)/3) e^{-\lambda E Q(E)/3}) \right] \, dE}{\int_{\text{spectrum}} S(E) \left[ 1 - e^{-\lambda E Q(E)/3} \right] \, dE}
\]

(13)

where \( \varepsilon_D \) is the fraction-corrected double efficiency, \( \varepsilon_d \) is the fraction-uncorrected double efficiency calculated with the correct figure of merit \( \lambda \) for a two photomultiplier tube system, and \( \varepsilon_T \) is the activity determined without fraction correction. For the TDCR method, \( \varepsilon_D < \varepsilon_T \) or \( \varepsilon_d < \varepsilon_T \). Taking the \(^3\)H source counted with \( f = 50\% \) as an example, if the true figure of merit for a liquid scintillator is 0.7932 e/keV, the experimental \( R_f \) value for \( f = 50\% \) would be 0.3682 which can be calculated using formula (9) with \( \lambda = 0.7932 \) e/keV and \( f = 0.50 \). The fraction-corrected detection efficiency of logical sum of double coincidence and triple coincidence, respectively, without fraction correction. For the TDCR method, \( \varepsilon_D < \varepsilon_T \) or \( \varepsilon_d < \varepsilon_T \). Taking the \(^3\)H source counted with \( f = 50\% \) as an example, if the true figure of merit for a liquid scintillator is 0.7932 e/keV, the experimental \( R_f \) value for \( f = 50\% \) would be 0.3682 which can be calculated using formula (9) with \( \lambda = 0.7932 \) e/keV and \( f = 0.50 \). The fraction-corrected detection efficiency of logical sum of double coincidence \( \varepsilon_D \) is 0.3944 calculated using formula (7) with \( \lambda = 0.7932 \) e/keV and \( f = 0.50 \). If the fraction-uncorrected TDCR calculation formula is used by letting the right hand side of formula (9) (with \( f = 0 \)) equal to 0.3682, the figure of merit would reduce to 0.5770 e/keV. Using this incorrect figure of merit to calculate the detection efficiency of logical sum of double coincidence \( \varepsilon_D \), the value obtained would be 0.4010. The activity would then be reduced by 1.65% according to formula (11). Such calculations were performed for \(^3\)H, \(^{63}\)Ni, \(^{14}\)C and \(^{90}\)Sr/\(^{90}\)Y for \( f = 0 \) to 0.5 and the discrepancy in activity as a function of the fraction of the rejection is shown in Fig. 3.

The influence of the rejection can be also found in the similar way for the CIEMAT/NIST method

![Fig. 3. Calculated discrepancies in activities, when no fraction correction is made, as a function of rejected fraction of the single photoelectron peak with TDCR method for \(^3\)H, \(^{63}\)Ni, \(^{14}\)C and \(^{90}\)Sr/\(^{90}\)Y.](image_url)
was Ultima Gold cocktail. These samples were equivalently type of 20 ml liquid scintillation vials. The scintillator used had volume of 10 ml contained in the ordinary commercial method.

samples in order to investigate the influence of changing 0.5062 e/keV is used to calculate the double efficiency for quenching standard would be reduced to 0.5062 e/keV. If a particular chemical quenching state by measuring a $^3$H nuclide source of interest.

Fig. 4. Calculated discrepancies in activities, when no fraction correction is made, as a function of rejected fraction of the single photoelectron peak with CIEMAT/NIST method for $^{63}$Ni, $^{14}$C and $^{90}$Sr/$^{90}$Y.

where $S(E)$ is the beta spectrum of the $^3$H quenching standard that has the same chemical quenching state as the nuclide source of interest.

For example, with rejection of 50% of the single photoelectron peak and no modification being made to the model, the figure of merit of 0.7932 e/keV obtained for a particular chemical quenching state by measuring a $^3$H quenching standard would be reduced to 0.5062 e/keV. If 0.5062 e/keV is used to calculate the double efficiency for $^{63}$Ni source in the same quench state without correction, the efficiency would be 0.6727. Comparing with the efficiency of 0.6834 calculated using the correct figure of merit 0.7932 e/keV and fraction-corrected efficiency calculation formula (6), an overestimation of activity by 1.56% is found. Such calculations were performed for $^{63}$Ni, $^{14}$C and $^{90}$Sr for $f = 0$ to $f = 0.5$ and the discrepancy in activity as a function of the fraction of rejection is shown in Fig. 4.

The opposite trend of the effect is seen in Figs. 3 and 4. Overestimation in activity for CIEMAT/NIST method is because of that the method uses $^3$H as a tracer and the nuclide of interest is of higher energy than the tracer. The fractional rejection results in larger reduction in the figure of merit for $^3$H than the nuclide.

4. Results and discussion

The activities of $^3$H, $^{63}$Ni, $^{14}$C and $^{90}$Sr/$^{90}$Y samples were 1765.4 Bq ± 1.1%, 3102.6 Bq ± 1.1%, 8471.6 Bq ± 0.6% and 8909.6 Bq ± 0.4%, respectively, determined using the measurement data obtained from zero discrimination with the TDCR method. For $^3$H and $^{63}$Ni, the dominant contribution to uncertainty was from the $kB$ value. For $^{14}$C and $^{90}$Sr/$^{90}$Y, the uncertainty in the $kB$ value and the standard deviation of the experimental TDCR and logical sum of double coincidence count rate were taken into account in the uncertainty evaluation. The sensitivity coefficients (or partial derivatives of the efficiency function) for each contributor to uncertainty were calculated numerically [12].

With the TDCR method, when approximately 25% and 50% of the single photoelectron peak was rejected, if no fraction-correction was made, the activities were found to be underestimated by 2.6% and 3.3% for $^3$H, 1.3% and 2.8% for $^{63}$Ni, 0.6% and 0.5% for $^{14}$C, and 0.4% and 0.3% for $^{90}$Sr/$^{90}$Y, respectively. The experimental results showed a much larger influence by the rejection than theoretical calculation. This was due to that the calculation model simulating the nearly perfect situation and the experimental data having uncertainties.

The results of both theoretical calculation and experiment also showed that, as one can predict, the influence to low-energy beta emitters was more significant than to high energy ones. This is easily understood in that, firstly the figure of merit has more influence on the detection efficiency for low-energy beta emitters than high energy ones in the range of $\lambda > 0.3$ e/keV. Secondly, for low-energy beta emitters, the single photoelectron peak contains a high proportion of total counts, while the majority of counts for
high-energy beta emitters are situated in the multiple photoelectron peaks.

The CIEMAT/NIST method was applied to the measurement data obtained from the TDCR system. The true figure of merit was obtained from the experimental detection efficiency of logical sum of double coincidence for the $^3$H sample using formula (7) with zero discrimination ($f = 0$) and found to be 0.7932 e/keV. 0.7932 e/keV was then used to determine the detection efficiency of logical sum of double coincidence for $^{63}$Ni, $^{14}$C and $^{90}$Sr/$^{90}$Y. With 25% and 50% of rejection, when no fraction-correction was undertaken, the figure of merit found from the $^3$H sample was 0.6282 and 0.5394 e/keV respectively. Using these figures of merit to determine the detection efficiency of logical sum of double coincidence and then activity for $^{63}$Ni, $^{14}$C and $^{90}$Sr/$^{90}$Y, it was found that the discrepancy in activity was within the uncertainty range for all these samples. In other words, negligible discrepancy was given by the experimental results even for the low-energy nuclide, $^{63}$Ni. However, the theoretical calculations showed that the CIEMAT/NIST method would overestimate the activity if no correction was made. Different results given by the theoretical calculations and experimental measurements are perhaps due to that the three-photomultiplier tube system was used for the investigation and the error caused in the figure of merit due to the same rejection was smaller for a three-photomultiplier tube system than for a two-photomultiplier tube system, as shown in Fig. 2(a), and the detection efficiency of logical sum of double coincidence was less sensitive to the figure of merit than the double efficiency in the two-photomultiplier tube system. The last assertion can be proved by comparing the partial derivatives $\frac{\partial \varepsilon_D}{\partial \lambda}$ from Eqs. (6) and (7) with $f = 0$. However, to make it simple, the comparison can be made on the detection efficiency for a single electron. Then, for the double efficiency for a two-photomultiplier tube system, $\varepsilon_d = (1 - e^{-\frac{E\varepsilon Q}{kB}})$, we have

$$\frac{\partial \varepsilon_D}{\partial \lambda} = EQ(E)e^{-\frac{E\varepsilon Q}{kB}} \left(1 - e^{-\frac{E\varepsilon Q}{kB}}\right) = EQ(E)P_{1,2}(0/m)R_{1,2} \quad (15)$$

where $P_{1,2}(0/m)$ and $R_{1,2}$ are the probability of detection of 0 photons and the detection efficiency for a single photomultiplier tube in a two-photomultiplier tube system respectively. For the detection efficiency of logical sum of double coincidence for a single electron, $\varepsilon_D = 3(1 - e^{-\frac{E\varepsilon Q}{kB}})^2 - 2(1 - e^{-\frac{E\varepsilon Q}{kB}})^3$, we have

$$\frac{\partial \varepsilon_D}{\partial \lambda} = 2EQ(E)e^{-\frac{E\varepsilon Q}{kB}} \left(1 - e^{-\frac{E\varepsilon Q}{kB}}\right) = 2EQ(E)P_{1,3}(0/m)^2R_{1,3} \quad (16)$$

where $P_{1,3}(0/m)$ and $R_{1,3}$ are the probability of detection of 0 photons and the detection efficiency for a single photomultiplier tube in a three-photomultiplier tube system respectively. It is obvious that $\frac{\partial \varepsilon_D}{\partial \lambda} < \frac{\partial \varepsilon_d}{\partial \lambda}$.

The activities versus the experimental TDCR for $^3$H and $^{63}$Ni samples are plotted in Figs. 5 and 6 for $f = 0\%$, 25% and 50% for $kB = 0.012$ with no fraction-correction being made for $^3$H, with and without fraction correction being made for $^{63}$Ni. It can be seen that not only do the activities diverge from the true value if no fraction-correction is made, the regression lines also become positively sloped except for $^3$H in the case of $f = 50\%$ where the slope of the regression line is negative. In fact, for all $kB$ value ranged from 0.007 to 0.015, the regression lines are sloped in the same direction for the same rejection, which is not shown in Figs. 5 and 6. This has not been observed in the $^{14}$C and $^{90}$Sr/$^{90}$Y samples where the regression line remained almost unchanged with or without correction. The sloped regression lines indicate that the calculated efficiency is not satisfactorily fitted to the experimental one and the correct
After the fraction correction was applied to $^{63}$Ni, the regression lines were resumed to have zero slopes giving a constant activity over the range of experimental TDCR values and the activity determined was the same as the true value within the uncertainty range. However, the correction could not successfully be applied to $^3$H. This is because of that the experimental TDCR over the entire range for $^3$H, after 25% and 50% rejection, becomes too low to be corrected. It was pointed out by Cassette [13] that TDCR ≤ 0.3, roughly corresponding to an expected mean number of 1 photoelectron per photomultiplier, was a threshold below which the TDCR model failed.

5. Conclusion

Although the experimental results showed much higher percentage of discrepancy in the figure of merit and activity for the TDCR method than theoretical calculations and lower for the CIEMAT/NIST method when a fraction of the single photoelectron peak is rejected by setting the discrimination level without making any fraction-correction, the following conclusions can be drawn with consideration of the experimental uncertainties:

(1) For low-energy beta emitters such as $^3$H and $^{63}$Ni, any rejection could introduce an error in the activity determination.

(2) For the middle energy range of beta nuclides such as $^{14}$C, the influence of up to 25% of rejection is minimal. With more than 25% of rejection, the discrepancy in activity is not negligible.

(3) For high-energy beta emitters such as $^{90}$Sr/$^{90}$Y, rejection of up to 50% of the single photoelectron peak does not cause any significant deviation in the activity.

(4) If no fraction correction is made for the rejection, one would underestimate the activity with the TDCR method and overestimate the activity with the CIEMAT/NIST method according to the theoretical calculations.

(5) The discrepancy in activity can be eliminated by applying the fraction correction to the efficiency calculation. However, if the experimental TDCR is approximately less than 0.3, the TDCR model itself failed and the correction cannot be applied.

For the TDCR method, whether existing a zero-gradient regression line of the plot of the activities versus the experimental TDCR could be a measure of negligible or significant discrimination level set for the measurement system.

References

Chapter Four

Development of activity standard for $^{90}$Y microspheres

L. Mo$^{a,b}$, B. Avci$^c$, D. James$^c$, B. Simpson$^d$, W.M. Van Wyngaardt$^d$, J.T. Cessna$^e$, C. Baldock$^b$

$^a$ Australian Nuclear Science and Technology Organisation, New Illawarra Road, Lucas Heights, NSW 2234, Australia

$^b$ Institute of Medical Physics, School of Physics, University of Sydney, NSW 2006, Australia

$^c$ SIRTeX Medical Limited, Unit F6, Parkview, 16 Mars Road, Lane Cove, NSW 2066, Australia

$^d$ CSIR National Metrology Laboratory, 15 Lower Hope Road, Rosebank, Cape Town 7700, South Africa

$^e$ National Institute of Standards and Technology, Gaithersburg, MD 20899, U.S.A

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Author’s contribution

L. Mo (candidate)
Developed experimental procedure, carried out experiment, analysed data and wrote manuscript.

B. Avci
Carried out experiment.

D. James
Contributed to the manuscript editing.

B. Simpson
Carried out experiment and edited manuscript.

F. Morris
Carried out experiment.

J.T. Cessna
Carried out experiment.

C. Baldock
Edited manuscript.
Development of activity standard for $^{90}\text{Y}$ microspheres

L. Mo$^{a,b,*}$, B. Avcic$^c$, D. James$^e$, B. Simpson$^d$, W.M. Van Wyngaardtd, J.T. Cessna$^e$, C. Baldock$^b$

$^a$Australian Nuclear Science and Technology Organisation, New Illawarra Road, Lucas Heights, NSW 2234, Australia
$^b$Institute of Medical Physics, University of Sydney, NSW 2006, Australia
$^c$SIRTeX Medical Limited, Unit F6 Parkview, 16 Mars Road, Lane Cove, NSW 2066, Australia
$^d$CSIR National Metrology Laboratory, 15 Lower Hope Road, Rosebank, Cape Town 7700, South Africa
$^e$National Institute of Standards and Technology, Gaithersburg, MD 20899, USA

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Abstract

$^{90}\text{Y}$ microspheres are important therapeutic radiopharmaceuticals used in the treatment of liver cancer through a process known as selective internal radiation therapy. SIR-spheres$^R$ is a radiopharmaceutical product that is comprised of $^{90}\text{Y}$ microspheres suspended in sterile, pyrogen-free water for injection into patients. It is necessary to establish for the SIR-spheres$^R$ production the capability of accurately measuring the activity of this product to a traceable national measurement standard. An activity standard for SIR-spheres$^R$ was developed from a standard for $^{90}\text{Y}$ solution, employing a highly quantifiable chemical digestion process. Calibration factors for the manufacturer’s ionisation chambers were determined for 1 and 5 ml of the SIR-spheres$^R$ product placed in Wheaton vials, for both 34% and 44% of $^{90}\text{Y}$ microsphere concentration.

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Keywords: $^{90}\text{Y}$; Solution; Microspheres; SIR-spheres$^R$; Activity measurement; Chemical digestion; Ionisation chamber calibration factor

1. Introduction

$^{90}\text{Y}$ microspheres are therapeutic radiopharmaceuticals used in the treatment of liver cancer (Houle et al., 1989). Following implantation via a catheter in the hepatic artery, $^{90}\text{Y}$ microspheres become embolised in the microvasculature of liver cancer where they irradiate the tumour by a process known as Selective Internal Radiation Therapy (Stubbs and Cannan, 2002). This results in the destruction of the tumour, while leaving most of the healthy liver tissue relatively intact.

SIR-spheres$^R$ is a radiopharmaceutical product comprised of $^{90}\text{Y}$ microspheres suspended in sterile, pyrogen-free water for injection into patients. They are produced regularly by Australian Nuclear Science and Technology Organisation (ANSTO) Radiopharmaceuticals and Industrials (ARI) and exported by SIRTeX, an Australian Company, to the international market. In order to meet the traceability requirement of the end users and provide confidence in the product, there is a need to standardise the product and establish a traceable chain of radioactivity measurement.

In the SIR-spheres$^R$ product, the microspheres are non-biodegradable plastic beads, have a density of

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1.0 g/ml and a diameter of approximately 35 μm. 90Y used was the nuclear reactor-produced radionuclide. The concentration of the microspheres is approximately 40% and patient doses of SIR-spheres® product are usually prepared in a Wheaton vial by ARI with an activity of 3 GBq in a volume of 5 ml. The activity administrated to each patient is approximately between 1 and 2.5 GBq for metastatic patients, and up to approximately 4.5 GBq for hepatocellular carcinoma patients. The activity measurement of each vial is performed in an ionisation chamber located in the ARI 90Y production area. The make and model of this ionisation chamber is unknown. The activity measured in this chamber is checked with a TPA ionisation chamber (Sharpe and Wade, 1951) and a Vinten ionisation chamber (NE Technology, 1991) located in the ARI Quality Control (QC) Section, using 1 ml SIR-spheres® sample dispensed from the same stock of SIR-spheres® as the product. In this paper these three chambers will be noted as the ARI production chambers, ARI QC TPA and ARI QC Vinten chambers, respectively. Fig. 1 shows 1 and 5 ml SIR-spheres® contained in Wheaton vials.

The aim of this work was to establish for the ARI SIR-spheres® manufacture and quality control process the capability to reliably measure the activity of SIR-spheres® with traceability to national measurement standards. This paper describes how the process was accomplished.

2. Method

90Y decays by pure beta emission to 90Zr with an end point energy of 2.28 MeV. The half-life of 90Y is 64.053 ± 0.020 h (Kossert and Schrader, 2004). The triple-to-double coincidence ratio (TDCR) method (Simpson and Meyer, 1994; Zimmerman and Ratel, 2005) and 4πβ high-pressure proportional counting system (Woods et al., 1996) were also used for the standardisation of 90Y.

In this work, primary and secondary standardisations were first performed on a 90Y solution. Through chemical digestion of the SIR-spheres®, a secondary standard factor for the 90Y solution was then converted into a secondary standard factor for the SIR-spheres®. Finally, ARI production chamber and ARI QC TPA and Vinten chambers were calibrated for the SIR-spheres®.

The ionisation chamber calibration factors were determined using samples with two different microsphere concentrations to examine the effect of microsphere concentration on the calibration factor.

Experimental uncertainties were evaluated in accordance with the ISO GUM (International Organisation for Standardisation, 1993) and are quoted here as standard uncertainties which have coverage factor k = 1 providing a confidence level of 68% unless otherwise specified.

2.1. Primary and secondary standardisation of 90Y solution

90Y in 0.1 M sulphuric acid solution was produced at ARI. The sulphuric acid was used to dissolve the active yttrium oxide powder. Two sample sets were prepared from this stock solution. One sample set with volume of 5 ml was used for primary standardisation at the National Institute of Standards and Technology (NIST) in USA using the CIEMAT/NIST method of liquid scintillation efficiency tracing with 3H to determine the absolute activity concentration (in MBq/g) of the stock solution. The other sample set with volumes of 1.5 and 5 ml was used to determine the current response (in pA/g) of the ANSTO Secondary Standard ionisation chamber. The volumes are nominal values. The exact mass of the solution was determined by weighing the vial together with the bung and cap before and after dispensing. The calibration factor (in pA/MBq) for the ionisation chamber was then calculated from the ratios of the results of the two sets of measurements.

The same procedure was carried out on another occasion, but this time the primary standardisation was performed at the Council for Scientific and Industrial Research–National Measurement Laboratory (CSIR-NML) in South Africa using the TDCR method. These two batches were unrelated in origin and prepared at the different times. On both occasions, the solutions were checked for photon-emitting impurities with a germanium detector. Confidence in the measurements made by NIST and the CSIR-NML was established through participation in an international comparison of activity.
measurements of $^{90}\text{Y}$ (Zimmerman and Ratel, 2005). Comparable results were obtained with those of the other participating laboratories, the results agreeing with the average value to better than 0.15% and within one standard deviation of the inter-comparison mean value.

The ANSTO Secondary Standard ionisation chamber is a TPA ionisation chamber. Volumes of 1.5 and 5 ml of $^{90}\text{Y}$ solution in Wheaton vials were calibrated for the ionisation chamber using the following formula that includes the correction for photon emitting impurities:

$$C_{90\text{Y}} = I_s A_{90\text{Y}} / C_{0\text{imp}},$$

where $C_{90\text{Y}}$ (pA/MBq) is ionisation chamber calibration factor for $^{90}\text{Y}$, $I_s$ (pA) is the sample ionisation current measured in the ionisation chamber and corrected for background current and instrument drift (using Ra-226 reference sources), $A_{90\text{Y}}$ (MBq) is the $^{90}\text{Y}$ activity of the sample, $R_i$ is the activity ratio of impurity to $^{90}\text{Y}$ and $C_{i\text{imp}}$ (pA/MBq) is the calibration factor for the impurity.

### 2.2. Deriving the calibration factor for 1 ml SIR-spheres® from the secondary standard for $^{90}\text{Y}$ solution

One milliliter SIR-spheres® samples with two different microsphere concentrations (34% and 44% in volume) were prepared in Wheaton vials at ARI. The SIR-spheres® stock was stirred for approximately 15 min, before dispensing was done, to ensure the homogeneity. Two samples were prepared for each concentration. These samples were labelled $^{90}\text{Y}$-m-1 and $^{90}\text{Y}$-m-2 for the concentration of 34% and $^{90}\text{Y}$-m-3 and $^{90}\text{Y}$-m-4 for 44%. They were checked for photon-emitting impurities with a germanium detector.

Fig. 2 illustrates the procedure for deriving the ionisation chamber calibration factor for 1 ml SIR-spheres® sample.

#### 2.2.1. Ionisation current measurement of 1 ml SIR-spheres®

As shown in the Fig. 2, the 1 ml SIR-spheres® samples ($^{90}\text{Y}$-m-1, $^{90}\text{Y}$-m-2, $^{90}\text{Y}$-m-3 and $^{90}\text{Y}$-m-4) were first measured in the ANSTO Secondary Standard and ARI QC TPA and ARI QC Vinten chambers to obtain current responses. To eliminate the effect of movement of microspheres on the current measurement, the samples were allowed to completely settle with the microspheres resting at the bottom of the vial, before measurements were undertaken in the ionisation chambers, ensuring a reproducible geometry. It took about 15 min for complete settling after the cessation of the agitation.

#### 2.2.2. Chemical digestion

The secondary standard developed for $^{90}\text{Y}$ in solution form is not suitable for the determination of the activity of SIR-spheres® due to the different sensitivity (National Council on Radiation Protection and Measurements, 1985) of an ionisation chamber to $^{90}\text{Y}$ in these two different forms. In order to transfer the secondary...
standard factor for $^{90}\text{Y}$ solution to the SIR-spheres®, a process of chemical digestion was employed.

As shown in the Fig. 2, after being measured in the various ionisation chambers, SIR-sphere® samples then underwent chemical digestion. The sample vial with reagents added in was placed inside a 50 ml glass beaker containing approximately 20 ml of water. The vials were crimp-sealed. A hotplate was used to heat the water to between 60 and 85 °C to increase the rate of digestion. The reaction of microspheres with the reagents caused vapours to be produced. A vial containing 0.5 ml of 0.1 M H$_2$SO$_4$ solution was set up to collect the vapour from the sample digestion through a plastic tube. Both ends of the tube were connected to a long needle. One needle was plugged inside the sample vial, suspended above the solution. The other needle was plugged into the H$_2$SO$_4$ solution in the vapour collection vial, so any $^{90}\text{Y}$ atoms that were released could be dissolved. The collection of vapour also reduced pressure build-up in the closed vial due to heat and reaction. The activity of the solution in the vapour collection vial was measured in a 4$\pi$β proportional counter. This activity (called activity loss), if significant, would be added to the sample activity. After the digestion, the sample volume increased to 2.2 ml as a result of the addition of chemical agents for digestion of the SIR-spheres®. The volume of each sample was then made up to 5 ml with the addition of 0.1 M HCl. Finally the digested SIR-spheres® were measured in the ANSTO Secondary Standard ionisation chamber and the activity determined using the calibration factors for a 5 ml solution. Further details of the digestion process cannot be disclosed for commercial reasons.

After chemical digestion, four 5 ml sample solutions for ionisation chamber measurements and four vapour collection solutions for quantification of activity loss during chemical digestion were produced from the original four samples $^{90}\text{Y}$-m-1, $^{90}\text{Y}$-m-2, $^{90}\text{Y}$-m-3 and $^{90}\text{Y}$-m-4.

The chemical digestion process described above was carried out with non-active SIR-spheres® samples before the active samples. The non-active digested SIR-spheres® were examined under a microscope with a magnification of ×20 to ensure that the digestion process was appropriate and the digestion was completed.

2.2.3. Measurement of activity lost in the chemical digestion

From each of the four vapour collection solutions, a 4$\pi$ counting source was produced by depositing known amount of aliquots onto a VYNS film (Wyllie, 1989). The VYNS counting sources were then measured in the 4$\pi$β gas-flow proportional counter, the counting gas being argon containing 10% methane. With the beta channel main amplifier in its normal (not overloaded) state, the beta particle counting efficiencies achieved previously were 97% for $^{198}\text{Au}$ (maximum beta energy of 1372.5 keV) (Reinhard et al., 2003) and 96% for $^{60}\text{Co}$ (maximum beta energy of 317.9 keV) (Mo et al., 2004). The $^{90}\text{Y}$ beta emissions have a comparatively high maximum energy of 2283.9 keV. It is thus reasonable to assume the beta particle counting efficiencies for $^{90}\text{Y}$ to be 98%. The uncertainty in the activity measurement due to this assumption was estimated to be less than 2%.

Non-extendable dead times of 8.00 ± 0.01 µs were measured. Each source was counted for ten intervals of 300 s duration. Counting data were collected on two subsequent days for each source. The observed count rates for each sample were corrected for background and dead time using Wyllie’s equation (Wyllie, 1987).

2.2.4. Determination of calibration factor for 1 ml SIR-spheres®

After chemical digestion, a 1 ml SIR-spheres® sample became a 5 ml solution where activity was then able to be determined using the secondary standard calibration factor for the 5 ml solution. The calibration factor for 1 ml SIR-spheres® was calculated as a ratio of the ionisation current of a 1 ml SIR-spheres® sample before digestion to the total activity of the sample. The total activity includes the digested SIR-spheres® sample activity plus the activity loss during chemical digestion, if the loss is significant.

2.3. Determination of calibration factor for 5 ml SIR-spheres®

As described in the Introduction, patient doses of SIR-spheres® are prepared in a Wheaton vial in a volume of 5 ml. The activity of the patient dose is measured in the ARI production chamber. Therefore, it was necessary to calibrate this chamber for 5 ml SIR-spheres®. The calibration factors for 5 ml SIR-spheres® were also derived for the ANSTO Secondary Standard, ARI QC TPA and Vinten chambers. Due to the difficulties of completely digesting 5 ml SIR-spheres without adding high volumes of reagents and still keeping the sample volume measurable in an ionisation chamber, the calibration factors were derived from the secondary standard factor for 1 ml SIR-spheres®.

Two 1 ml and two 5 ml SIR-spheres® samples with microsphere concentration of approximately 43% (by volume) were prepared from the same stock at ARI. The samples were checked for photon-emitting impurities with a germanium detector.

Two 5 ml SIR-spheres® samples were measured in all the ionisation chambers so as to obtain the ionisation current. Two 1 ml SIR-spheres® samples were measured in the ANSTO Secondary Standard ionisation chamber in order to determine the activity concentration of the SIR-spheres® stock, from which the activities of the 5 ml
SIR-spheres® samples were calculated. The calibration factor for 5ml SIR-spheres® was then determined by dividing the SIR-spheres® sample current by its activity. Due to practicality, the 5ml samples were measured in the ARI production chamber immediately after dispensing and before the microspheres settled at the bottom. Five repeated measurements were made in 2 min.

3. Results and discussion

3.1. Primary and secondary standardisation of 90Y solution

The 88Y impurity was detected in both batches of the primary standardisation solutions, with 0.00046% for the CIEMAT/NIST method batch and 0.00026% for the TDCR method batch. The TDCR measurement result was checked against the CSIR-NML ionisation chamber, for which a calibration figure had been obtained during the international intercomparison. Agreement to better than 1% was obtained, giving confidence in the measurements.

The response of the ionisation chamber to 90Y was low since the chamber measured almost entirely low-energy bremsstrahlung produced by the interactions of the beta particles mainly with the solution and vial. 88Y emits high-energy gamma rays in the range 484–3218 keV. The predominant photons emitted have energies of 898 and 1836 keV with emission probabilities of 94% and 99%, respectively. The calibration factor of 88Y for the ANSTO Secondary Standard ionisation chamber is 65 pA/MBq which is nearly 300 times more than that of 90Y. However, the content of 88Y impurity was too low in respect of 90Y activity to have a significant effect on the calibration factor.

The calibration factors for the ANSTO Secondary Standard ionisation chamber derived from the primary standardisations using Eq. (1) were 0.238 pA/MBq ± 0.83% for the 1.5 ml 90Y solution and 0.218 pA/MBq ± 0.74% for the 5 ml 90Y solution. The calibration factors decreased with increasing volume, in agreement with the experiments of Coursey et al. (1993).

3.2. Determination of calibration factor for 1 and 5 ml SIR-spheres

The activity level for photon emission impurities found in samples 90Y-m-1, 90Y-m-2, 90Y-m-3 and 90Y-m-4 were below the detection limit of the germanium detector. The measured ionisation currents of these samples before digestion and their activities determined using the secondary standard factor for 5 ml solution are listed in Table 1. The uncertainty components for the ionisation currents and activities are given in Table 2.

The plastic tubes connecting two vials for the collection of vapour from digestion were examined by a surface contamination monitor and negligible count rate was observed. The activity loss for each 1ml SIR-spheres® sample during chemical digestion is tabulated in Table 3.

From Table 3, it can be seen that the activity loss was less than 0.006% of the activity of the SIR-spheres® sample before digestion. Therefore, the activity loss was considered negligible. However, 0.006% was assigned as a B type uncertainty component (as shown in Table 2).

The calibration factors for 1ml SIR-spheres® in a Wheaton vial for each ionisation chamber are listed in Table 4. The uncertainties are combined uncertainties of the components due to the ionisation currents and activities shown in Table 1.

There was no obvious change in the ionisation current for the first and last measurement for the same unsettled sample, measured in the ARI production ionisation chamber. It might be because of that the whole set of measurements were completed in 2 min that was not long enough for the microspheres to move greatly to affect the current. The calibration factors for 5ml SIR-spheres® in a Wheaton vial for each ionisation chamber are listed in Table 5. Although 0.00006% of 88Y impurity was detected in this batch, it was too low to make impurity corrections.

The factor for 5ml SIR-spheres® derived in this way might result in a larger uncertainty because the SIR-spheres® stock might not be very homogeneous although the stirrer was used. This can be seen from the standard deviation, which was found to be 1%, of

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<thead>
<tr>
<th>Sample</th>
<th>Ionisation current (pA) before chemical digestion</th>
<th>Activity (MBq)²</th>
</tr>
</thead>
<tbody>
<tr>
<td>90Y-m-1</td>
<td>350.4 ± 0.88% ANSTO Secondary Standard</td>
<td>1452 ± 1.2%</td>
</tr>
<tr>
<td>90Y-m-2</td>
<td>284.7 ± 1.7% ARI TPA</td>
<td>1445 ± 1.2%</td>
</tr>
<tr>
<td>90Y-m-3</td>
<td>100.5 ± 1.6% ARI Vinten</td>
<td>2108 ± 1.2%</td>
</tr>
<tr>
<td>90Y-m-4</td>
<td>415.2 ± 1.7%</td>
<td>2093 ± 1.2%</td>
</tr>
</tbody>
</table>

²The activity quoted in this table does not include the activity loss during chemical digestion.
the specific ionisation current value (pA/g) for the two 1ml and two 5ml SIR-sphere\textsuperscript{®} samples dispensed from the same stock. The microspheres tended to sink when the SIR-spheres\textsuperscript{®} stock was moved away from the stirrer for dispensing. The uncertainty components and evaluation for the calibration factors for 5ml

Table 2
Uncertainty components for the ionisation currents and activities of 1ml \textsuperscript{90}Y SIR-spheres\textsuperscript{®}

<table>
<thead>
<tr>
<th>Source of uncertainty</th>
<th>Relative standard uncertainty components (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>ANSTO Secondary Standard</td>
</tr>
<tr>
<td></td>
<td>Type A</td>
</tr>
<tr>
<td>Electrometer accuracy</td>
<td>0.14</td>
</tr>
<tr>
<td>ESDMa of sample current</td>
<td>0.027</td>
</tr>
<tr>
<td>Background</td>
<td>0.0004</td>
</tr>
<tr>
<td>Chamber drift correction</td>
<td>0.044</td>
</tr>
<tr>
<td>Chamber drift (no correction)</td>
<td></td>
</tr>
<tr>
<td>Non-linearity of chamber</td>
<td></td>
</tr>
<tr>
<td>\textsuperscript{90}Y decay correction</td>
<td></td>
</tr>
<tr>
<td>Calibration factor</td>
<td></td>
</tr>
<tr>
<td>Activity loss during digest</td>
<td></td>
</tr>
<tr>
<td>Combined uncertainty</td>
<td>0.88</td>
</tr>
</tbody>
</table>

\textsuperscript{a}Experimental standard deviation of the mean.

Table 3
Activity loss during chemical digestion

<table>
<thead>
<tr>
<th>Sample</th>
<th>Activity loss (kBq)</th>
<th>Activity loss/total activity</th>
</tr>
</thead>
<tbody>
<tr>
<td>\textsuperscript{90}Y-m-1</td>
<td>32.2±2%</td>
<td>0.002%</td>
</tr>
<tr>
<td>\textsuperscript{90}Y-m-2</td>
<td>18.5±2%</td>
<td>0.001%</td>
</tr>
<tr>
<td>\textsuperscript{90}Y-m-3</td>
<td>102.1±2%</td>
<td>0.005%</td>
</tr>
<tr>
<td>\textsuperscript{90}Y-m-4</td>
<td>133.2±2%</td>
<td>0.006%</td>
</tr>
</tbody>
</table>

Table 4
Ionisation chamber calibration factors for 1ml SIR-spheres\textsuperscript{®} in a Wheaton vial

<table>
<thead>
<tr>
<th>Geometry</th>
<th>Ionisation chamber calibration factors (pA/MBq)</th>
</tr>
</thead>
<tbody>
<tr>
<td>ANSTO Secondary Standard</td>
<td>ARI TPA</td>
</tr>
<tr>
<td>1 ml SIR-spheres\textsuperscript{®} in Wheaton vial with 34% (by volume) of \textsuperscript{90}Y microspheres</td>
<td>0.241±1.0%</td>
</tr>
<tr>
<td>1 ml SIR-spheres\textsuperscript{®} in Wheaton vial with 44% (by volume) of \textsuperscript{90}Y microspheres</td>
<td>0.242±1.0%</td>
</tr>
</tbody>
</table>

Table 5
Ionisation chamber calibration factors for 5 ml SIR-spheres\textsuperscript{®} in a Wheaton vial

<table>
<thead>
<tr>
<th>Geometry</th>
<th>Ionisation chamber calibration factors (pA/MBq)</th>
</tr>
</thead>
<tbody>
<tr>
<td>ANSTO Secondary Standard</td>
<td>ARI TPA</td>
</tr>
<tr>
<td>5 ml SIR-spheres\textsuperscript{®} in Wheaton vial with 43% (by volume) of \textsuperscript{90}Y microspheres</td>
<td>0.223±2.2%</td>
</tr>
</tbody>
</table>
SIR-spheres® were similar as described before, except for the additional components of the sample mass (0.70%) and inhomogeneity of the activity concentration of the SIR-spheres® stock (1.7%).

Considering the primary solution as having a 0% microsphere concentration, it can be seen that the calibration factors increase somewhat with increasing microsphere concentration, by about 1% for 1 ml and 2% for 5 ml when the concentration increases from 0% to 44%. The density of the microspheres and that of water are almost the same. The amount of bremsstrahlung generated and absorbed by microspheres and water should be similar. The factor could be considered for the small discrepancy is how the active substances are distributed in these two types of radioactive sources. In the microspheres source, the active substances are attached to the surface of microspheres; while in the solution source, they are uniformly distributed. Further study of this effect needs to be undertaken. For the same microsphere concentration however, the calibration factors decrease greatly with increasing volume, approaching 10% depending on ionisation chamber type, showing that the volume has a more significant effect than the microsphere concentration on the calibration factor.

4. Conclusion

The Secondary Standard ionisation chamber calibration factor for ⁹⁰Y solution may be accurately transferred to SIR-spheres® by employing a highly quantifiable chemical digestion method. The direct standard transfer is limited to a small quantity of SIR-spheres® of about 1 ml due to the difficulty of effectively digesting a large quantity of SIR-spheres® such as 5 ml. The calibration factor for 5 ml SIR-spheres® may be derived from the factor for 1 ml SIR-spheres®, although relatively large uncertainty may be introduced due to the inhomogeneity of activity concentration dispensed into 1 and 5 ml samples. Under this principle, the ARI production chamber and ARI QC TPA and Vinten chambers were calibrated for 1 and 5 ml SIR-spheres® in Wheaton vials. The capability for ARI to perform traceable activity measurements of the SIR-spheres® product was therefore developed.

Acknowledgments

The author would like to thank ARI for their cooperation with the sample preparation and arrangement of the experimental area. The author would also like to thank Ms C. Maloney, director of Safety and Radiation Science ANSTO, for her constant support of this work, Dr. D. Alexiev and Dr. M. Reinhard, ANSTO, for their useful suggestions in experimental techniques and Mr. J.B. Davies, ANSTO, for checking experimental data calculations.

References

Sharpe, J., Wade, F., 1951. TPA Mk III ionisation chamber. AERE EL/R806.
Wyllie, H.A., 1989. The preparation of radioactive sources with radioactivities of less than 110 kilobecquerels, ANSTO/M118.
Chapter Five

Activity determination of $^{198}$Au solid source using $4\pi\beta-\gamma$ coincidence counting corrected by Monte-Carlo calculation

L. Mo$^{a,b}$, H.Y. Wu$^a$, C. Baldock$^b$

$^a$Australian Nuclear Science and Technology Organisation, New Illawarra Road, Lucas Heights, NSW 2234, Australia

$^b$Institute of Medical Physics, School of Physics, University of Sydney, NSW 2006, Australia

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Author’s contribution

L. Mo (candidate)
Designed methodology and experiment, carried out experiment, analysed both experimental and Monte Carlo simulation data and wrote manuscript.

H.Y. Wu
Carried out Monte Carlo simulation and interpreted the simulation data.

C. Baldock
Edited manuscript.
Abstract—For the commissioning process of the OPAL nuclear reactor of the Australian Nuclear Science and Technology Organization (ANSTO), the thermal neutron flux is measured through the activity measurement of an activated Au wire, Au-Al (0.112% of Au) alloy wire and Au foil.

The absolute activities of $^{198}$Au in the form of Au wire, Al-Au wire and Au foil were determined using the conventional $4\pi\beta - \gamma$ coincidence-counting method. Monte Carlo simulation technique was employed to simulate the complicated absorption and attenuation processes of electrons and gamma photon interactions with the surrounding materials. The Monte Carlo calculated probabilities of escape beta particles, internal conversion electrons and photon-interaction generated photoelectrons and Compton electrons were used to determine the correction term of the coincidence equation. The corrections for the Au wire (length: 8.000 mm, radius: 0.064 mm), Al-Au wire (length: 7.690 mm, radius: 0.255 mm) and Au foil (thickness: 0.025 mm, radius: 3.000 mm) were found to be 5.2% $\pm$0.1%, 2.6% $\pm$0.1% and 4.2% $\pm$0.2% respectively. The study demonstrates that the Monte Carlo calculation for the correction term of the coincidence equation can be applied to the absolute activity determination of radionuclides with well-defined source geometries with an uncertainty of better than 1%.

Index Terms—$^{198}$Au, $4\pi\beta - \gamma$ coincidence-counting, activity, Monte Carlo simulation.

I. INTRODUCTION

THERMAL neutron flux measurement is commonly carried out through the activity measurements of $^{198}$Au which is induced by the neutron reaction $^{197}$Au(n, $\gamma$) $^{198}$Au. During the hot commissioning stage of the OPAL nuclear reactor at the Australian Nuclear Science and Technology Organization (ANSTO), three types of Au materials, namely pure Au wire (99.9971% of Au), Al-Au alloy wire (0.112% of Au) and pure Au foil (99.99% of Au) were used for the measurement of neutron flux distribution in the reactor core, irradiation facilities and neutron beam lines. This paper describes the work carried out on the determination of the absolute activity of $^{198}$Au in the form of Au wire, Al-Au alloy wire and Au foil.

$^{198}$Au has a simple decay scheme as shown in Fig. 1. It decays to $^{198}$Hg through 100% $\beta^-$ emission. The predominant $\beta$ branch (a branch) proceeds to the 411.802 keV excited state level of $^{198}$Hg, which promptly de-excites to ground state via emission of gamma ray $\gamma_1$ of energy 411.802 keV (intensity 95.54%). This $\beta$ branch has an emission probability of 98.986% and maximum energy of 960.4 keV. The second $\beta$ branch (b branch) proceeds to the 1087.687 keV excited state level of $^{198}$Hg, which promptly de-excites to ground state via emission of gamma rays $\gamma_2$ and $\gamma_3$ of energy 675.885 keV (intensity 0.806%) and 1087.687 keV (0.159%) respectively. This branch has much lower emission probability of 0.989% and maximum energy 284.5 keV. The very weak $\beta$ branch (c branch) proceeds to the ground state of $^{198}$Hg, and is usually ignored when absolute activity is measured. The half-life of $^{198}$Au is 2.6944 $\pm$0.0008 days [1].

The $4\pi\beta - \gamma$ coincidence-counting method [2] is an ideal method for standardizing such a nuclide. In the coincidence-counting system used in this work, a 4 $\pi$ gas-flow proportional counter was used for the event detection induced by $\beta$-particles. A 3 by 3 inch thallium activated sodium iodide NaI(Tl) scintillator was used for the detection of $\gamma$-rays.

For thick sources such as Au wires and foils, the problem of using the $4\pi\beta - \gamma$ coincidence-counting method is the source self-absorption and interaction of gamma photons with source material which generates appreciable amount of phototelectrons and Compton electrons. The detection efficiency of $\beta$-particles is less than 60%. For such low $\beta$-efficiency, the efficiency extrapolation method [3] is not suitable. The activity can only be determined by applying a correction term to the basic coincidence equation [2], [4]. The correction term is due to the detection of unwanted radiations and charged particles, including

1087.687 keV

$^{198}$Au

$^{198}$Hg

$\gamma_1$

$\gamma_2$

$\gamma_3$

411.802 keV

Fig. 1. Decay scheme of $^{198}$Au.
the second branch (b branch) \( \beta \)-particles, \( \gamma_2 \) (675 keV) and \( \gamma_3 \) (1087 keV) transitions, by the \( \beta \)- and \( \gamma \)-detectors.

The \( \gamma \)-ray sensitivity \( \varepsilon_{\gamma} \) of a \( \beta \)-detector depends on two separate factors: 1) the probability \( p_e \) of \( \gamma \)-ray interactions with the source material, gas and wall of the 4\( \pi \) proportional counter to produce the photoelectron and Compton electrons and 2) the probability \( P_c \) of so produced electrons escaping into the sensitive region of the counting gas. \( \varepsilon_{\gamma} \) can be evaluated by summing up the product of these two probabilities, i.e., \( \sum P_e P_c \), over all the available materials. The general approach for \( P_c \) and \( P_e \) is to compute the value using mass absorption coefficient and theoretical maximum range for monoenergetic electron [5]–[7].

In Axton’s work [5], the detection efficiencies were evaluated as following: 1) the efficiency \( \varepsilon_{\gamma, \beta} \) of a \( \beta \)-detector for the b branch \( \beta \)-particles by taking the \( \beta \)-particle escape probability as \( 1 - e^{-\lambda_t (t)} / \lambda_t \) and integrating the probability over the sample thickness \( t \); 2) the efficiency \( \varepsilon_e \) for the conversion electrons by using the theoretical maximum range for monoenergetic electrons; and 3) the efficiency \( \varepsilon_{\gamma, 2} \) and \( \varepsilon_{\gamma, 3} \) of \( \gamma \)-channel for the \( \gamma_2 \) and \( \gamma_3 \) photons by integrating the differential Compton absorption cross section over the energy \( E_1 \) to \( E_2 \) which are the limits of the window on the \( \gamma \)-channel.

Hargrove and Geiger [6] calculated \( \varepsilon_{\gamma, 2} \) and \( \varepsilon_{\gamma, 3} \) based on the published counting efficiencies for \( \gamma \)-rays. Parry and Urquhart [8] determined \( \varepsilon_{\gamma, 2} \) and \( \varepsilon_{\gamma, 3} \) experimentally, using the standardized sources of \(^{198}\text{Au}, \; ^{137}\text{Au}, \; ^{54}\text{Mn} \) and \(^{60}\text{Co} \).

Sazonova et al. [9] used a completely different approach. They studied the coincidence counting equations and found that the slope of the extrapolation function could be made zero for a certain ratio of efficiencies in the \( \gamma \)-channel. The ratio could be obtained by selecting a NaI(Tl) crystal of a certain thickness and absorbers of certain materials and thickness.

The above work was carried out on Au foils only and the corrections have achieved a certain degree of accuracy. The Monte Carlo simulation enables these corrections to be calculated with much higher accuracy. The application of Monte Carlo simulation to the 4\( \pi \beta \)-\( \gamma \) coincidence counting was carried out by Miyahara et al. [10]. In their work, the \( \beta \) energy spectra and self-absorptions of \(^{134}\text{Cs}, \; ^{56}\text{Fe}, \; ^{106}\text{Ru} \) and \(^{60}\text{Co} \) spherical particle sources were calculated by using the Monte Carlo method. The Monte Carlo simulation of all detection processes for \(^{134}\text{Cs} \) in a 4\( \pi \beta \)-\( \gamma \) coincidence counting system was performed recently by Takeda et al. [11]. Both of the work using the Monte Carlo simulation allowed the calculation of the efficiency extrapolation function.

In this work, the absolute activity of \(^{198}\text{Au} \) in the form of Au wire, Al-Au alloy wire and Au foil was measured in a 4\( \pi \beta \)-\( \gamma \) coincidence counting system with corrections determined by the Monte Carlo simulation. MCNP code (Monte Carlo N-Particle Transport code) Version 4B [12], was used to simulate the complicated absorption and attenuation processes of electrons and gamma photon interactions with the surrounding materials.

The application of MCNP in simulating electron transport and photon interaction with matter was validated by Takeda et al. [11] using the experimental data obtained from the 4\( \pi \beta \)-\( \gamma \) coincidence counting of \(^{134}\text{Cs} \).

II. METHODOLOGY

A. Source Preparation

The \(^{198}\text{Au} \) sources were obtained by neutron irradiation of Au wire, Al-Au alloy wire and Au foil at the ANSTO High Flux Australian Reactor (HIFAR). After decayed to an appropriate activity, the solid sources were placed onto 4\( \pi \) source mounts for coincidence counting. The source mount was VVNS (polyvinyl chloride-acetate copolymer) thin film [13] rendered conducting by the vacuum deposition of a thin layer of Au20% - Pd [14]. The surface density of the VVNS film was 15 \( \mu \)g/cm\(^2 \) and of coating 20 \( \mu \)g/cm\(^2 \). Three samples for each type of materials were prepared.

B. Coincidence Counting

A conventional 4\( \pi \beta \)-\( \gamma \) coincidence counting system was used. As shown in Fig. 2, the system consists of a 4\( \pi \) gas-flow proportional counter for detection of \( \beta \)-particles, a 3 by 3 inch thallium activated sodium iodide NaI(Tl) scintillator for detection of \( \gamma \)-rays and a series of electronic components for processing of pulse signals. The scintillator is located above the \( \beta \)-counter. The counter is filled with a 10% Methane in Argon (P-10) gas which flows through the counter at atmospheric pressure. The coincidence events are detected by a coincidence mixer connected to the outputs of the \( \beta \) and \( \gamma \) paralysis units.

As adopted in a normal procedure, the window in the \( \beta \) channel was left fully open so all of the beta particles were counted. The main amplifier in the channel was set in an overload state.

A window in the \( \gamma \) channel was set to gate the 411 keV photopeak. With this arrangement the counts recorded in the \( \gamma \)-channel were predominantly due to \( \gamma_1 \), plus multiple
Compton process events in which $\gamma_2$ and $\gamma_3$ deposited their partial energy to the gamma detector and the energy fell within the $\gamma$ window.

Each sample was counted for 10 times of 10 minutes duration for each counting. Repeated measurements were made for 14 days.

Ignoring the response of the extremely weak $\beta$ branch (c branch), for a source with a disintegration rate $N_0$, the count rate $N_\gamma$ detected by the $\beta$-detector with detection efficiency $\varepsilon_{\beta\gamma}$ and $\varepsilon_{\beta\alpha}$ for a and b branch $\beta$-particles is given by [(1) shown at the bottom of this page], where, $a$ and $b$ are the branch ratios of $a$ and $b$ branch $\beta$-particles, $\varepsilon_{\beta\gamma_1}$ $\varepsilon_{\beta\gamma_2}$ and $\varepsilon_{\beta\gamma_3}$ are the $\gamma$-ray sensitivities of the $\beta$-detector for $\gamma_1$, $\gamma_2$ and $\gamma_3$ photons, $\varepsilon_{\gamma_1\gamma_1}$, $\varepsilon_{\gamma_1\gamma_2}$, and $\varepsilon_{\gamma_1\gamma_3}$ are the efficiencies of the $\beta$-detector for conversion electrons from $\gamma_1$, $\gamma_2$ and $\gamma_3$ transitions, $d$ is the absolute $\gamma_2$ transition probability from $b$ branch $\beta^-$ emission, and $\alpha_1$, $\alpha_2$ and $\alpha_3$ are the total internal conversion coefficients for $\gamma_1$, $\gamma_2$, and $\gamma_3$ transitions respectively.

In (1),

$$g = \frac{1 - \varepsilon_{\beta\gamma}}{\varepsilon_{\beta\gamma}} \times \left[ b(1 - k) \left[ a + kd \frac{\varepsilon_{\gamma_1} + \varepsilon_{\gamma_2} + \varepsilon_{\gamma_3} - \varepsilon_{\gamma_1\gamma_2}}{1 + \alpha_2} \right] \right],$$

where, $k = (1 - \varepsilon_{\beta\gamma})/(1 - \varepsilon_{\beta\alpha})$. The detection efficiencies of Auger electrons and X-rays, produced by the rearrangement of electrons of an atom following internal conversion, are ignored due to the absorption of the low energy electrons in the solid source.

The gamma count rate $N_\gamma$ detected in the NaI detector with the efficiency $\varepsilon_{\gamma_1}$, $\varepsilon_{\gamma_2}$, and $\varepsilon_{\gamma_3}$ for $\gamma_1$, $\gamma_2$, and $\gamma_3$ photons whose energies fall within the $\gamma$ window is given by,

$$N_\gamma = N_0 \left[ \frac{a + d}{1 + \alpha_1} \varepsilon_{\gamma_1} + \frac{d}{1 + \alpha_2} \varepsilon_{\gamma_2} + \frac{(b - d)}{1 + \alpha_3} \varepsilon_{\gamma_3} \right] = N_0 \varepsilon_{\gamma_\beta} (1 + f), \quad (2)$$

where,

$$f = \frac{1}{a} \left( d + d \left\{ \frac{1 + \alpha_1}{1 + \alpha_2} \frac{\varepsilon_{\gamma_2}}{\varepsilon_{\gamma_1}} + \frac{b - d}{1 + \alpha_3} \frac{\varepsilon_{\gamma_3}}{\varepsilon_{\gamma_1}} \right\} \right).$$

The coincidence count rate $N_c$ registered in the coincidence mixer is given by [(3) shown at the bottom of this page], where,

$$h = \frac{1 - \varepsilon_{\beta\gamma}}{\varepsilon_{\beta\gamma}} \left[ f(1 - k) + \frac{1}{a} kd \frac{\varepsilon_{\gamma_1\gamma_2} + \varepsilon_{\gamma_1\gamma_3} + \varepsilon_{\gamma_2\gamma_3}}{1 + \alpha_2} \right].$$

The last two terms account for the $\gamma_2 - \gamma_1$ and $\gamma_2 - \gamma_3$ coincidences.

From (1), (2) and (3), we have

$$\frac{N_\beta N_\gamma}{N_c} = \frac{N_0 (1 + g)(1 + f)}{1 + f + h} = N_0 (1 + K), \quad \text{and}$$

$$\varepsilon_{\beta\gamma} = \frac{N_\gamma}{N_\beta} \varepsilon_{\gamma_\beta} (1 + f) \left\{ \frac{1}{1 + f + h} \right\} \quad \text{or}$$

$$K = \frac{g + gf - h}{1 + f + h}.$$

The activity $N_0$ can therefore be determined using (4). In the correction term $K$, $g$ is the correction component for complex decay scheme, $\gamma$-sensitivity of the $\beta$-detector and $\beta$-detector detection of conversion electrons; $f$ is the component for $\gamma$-detector detection of $\gamma_2$ and $\gamma_3$; and $h$ is the component for the unwanted coincidences. These values were obtained from the experiments and MCNP calculations, $\varepsilon_{\beta\gamma}$ was extracted from (5) and the ratios $\varepsilon_{\gamma_2}/\varepsilon_{\gamma_1}$ and $\varepsilon_{\gamma_3}/\varepsilon_{\gamma_1}$ were obtained experimentally using the standardized $4\pi$ sources of $^{198}\text{Au}$, $^{137}\text{Cs}$,
For the $\gamma$-rays, the Monte Carlo simulation accounted for the two major types of photon interaction with matter, namely photoelectric and Compton effects that occurred with $^{198}\text{Au}$. Pair production was not considered due to the very low intensity of 1087 keV photons (intensity 0.159%). The probability of a photon causing an emission of the photoelectrons and Compton electrons escaping from the source material and source mount into the counting gas were calculated. In both $\beta$- and $\gamma$-ray cases, the absorption of backscattered particles was considered. The generation of Auger electrons and X-rays were accounted, but regarded to be absorbed in the solid source.

A diagram of the $4\pi$ proportional counter and source mount modeled in the MCNP study is shown in Fig. 3. The counter is an aluminium cylinder with side wall thickness of 17.76 mm and top of 0.88 mm. In the middle of the cylinder is the stainless steel belt of 21.81 mm deep. The source mount was VYNS thin film coated with 20% Au and 80% Pd. The surface density of the VYNS film was 15 $\mu$g/cm$^2$ and of coating 20 $\mu$g/cm$^2$. The film was held by a circular brass ring with inner radius of 12.65 mm, outer radius 17.45 mm and thickness of 0.48 mm. The dimensions of the sources, source mount and gas simulated in the MCNP are listed in Table I. The source was placed in the centre of the source mount. A 10% Methane in Argon (99.997% Au) was used.

In order to evaluate the uncertainties due to the geometry variations, the MCNP calculations were also carried out for the sample with $\pm15\%$ deviation in length of the wires and diameter of the foil, and $\pm10\%$ in the diameter of the wires and thickness of the foil.

### D. Impurity Detection

Detection of photon emitting impurities was carried out using a GEM Series HPGe coaxial crystal detector (model No.: GEM-15200). The crystal dimension was 46.9 mm in diameter and 46.1 mm in length. The source was positioned at 100 mm away from the detector end cap (103 mm from crystal head).

### III. RESULTS AND DISCUSSION

#### A. Efficiency for $\beta$-Particles and Conversion Electrons

The MCNP calculated energy distributions of escape $\beta$-particles (into counting gas) of a and b branch for the pure Au wire...
are shown in Fig. 4. The initial energy distributions calculated theoretically for the \( \beta \)-particles of a and b branch are also shown in Fig. 4. The energy distributions for the Al-Au wire and Au foil, which are not shown in the figure, have the similar shape as Au wire.

It is assumed that, with the beta channel main amplifier set in an overloaded state, all \( \beta \)-particles or electrons entering into the gas will generate primary ionizing events and all the events will be detected by the \( \beta \)-detector. This reasonable simplification allows 1) the efficiency for \( \beta \)-particles to be calculated from the energy distribution of the escape \( \beta \)-particles; 2) the efficiency for conversion electrons to be calculated from the probability of the escape conversion electrons and 3) the \( \gamma \)-sensitivity of the \( \beta \)-detector to be calculated from the escape photoelectrons and Compton electrons. The MCNP calculated efficiencies \( \varepsilon_{\beta\gamma} \), \( \varepsilon_{\gamma\gamma} \), \( \varepsilon_{\gamma\gamma} \), \( \varepsilon_{\gamma\gamma} \), and \( \varepsilon_{\gamma\gamma} \) for the three types of samples are listed in Table II. For comparison, the measured efficiency \( \varepsilon_{\beta\gamma} \) is also listed in Table II.

From Table II, we can see that the calculated efficiency \( \varepsilon_{\beta\gamma} \) agreed with the measured efficiency for Au wire, whereas 10% lower for the Al-Au wire and 26% higher for the Au foil. The discrepancy for the Al-Au wire could be due to the error in the chemical composition which was provided by the supplier. For Au foil, the result shows that the MCNP has a limitation when dealing with electron multiple scattering in a very thin material. \( \varepsilon_{\beta\gamma} \) is the most influential parameter for the correction term. Its uncertainty dominates the combined uncertainty of the correction term. Fortunately, this parameter could be obtained experimentally with a small correction determined using MCNP calculations (see (5)). \( \varepsilon_{\beta\gamma} \) could not be obtained experimentally and therefore was determined by using the calculated value corrected for the difference between the experimental and calculated value based on \( \varepsilon_{\beta\gamma} \).

### Table II

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Efficiency value</th>
<th>Au wire</th>
<th>Al-Au wire</th>
<th>Au foil</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \varepsilon_{\beta\gamma} )</td>
<td>(28.9 ± 1.5)%</td>
<td>(45.1 ± 3.0)%</td>
<td>(74.6 ± 6.0)%</td>
<td></td>
</tr>
<tr>
<td>( \varepsilon_{\gamma\gamma} ) (measured)</td>
<td>(29.0 ± 0.1)%</td>
<td>(49.7 ± 0.1)%</td>
<td>(58.7 ± 0.4)%</td>
<td></td>
</tr>
<tr>
<td>( \varepsilon_{\gamma\gamma} )</td>
<td>(4.4 ± 0.4)%</td>
<td>(8.1 ± 0.8)%</td>
<td>(11.8 ± 1.2)%</td>
<td></td>
</tr>
<tr>
<td>( \varepsilon_{\gamma\gamma} )</td>
<td>(35.5 ± 2.3)%</td>
<td>(59.9 ± 4.4)%</td>
<td>(80.0 ± 8.0)%</td>
<td></td>
</tr>
<tr>
<td>( \varepsilon_{\gamma\gamma} )</td>
<td>(64.9 ± 2.0)%</td>
<td>(91.0 ± 3.1)%</td>
<td>(99.2 ± 6.0)%</td>
<td></td>
</tr>
<tr>
<td>( \varepsilon_{\gamma\gamma} )</td>
<td>(91.4 ± 0.5)%</td>
<td>(98.8 ± 0.6)%</td>
<td>(100.0 ± 1.0)%</td>
<td></td>
</tr>
</tbody>
</table>

### Table III

<table>
<thead>
<tr>
<th>Energy</th>
<th>Material</th>
<th>Au wire</th>
<th>Al-Au wire</th>
<th>Au foil</th>
</tr>
</thead>
<tbody>
<tr>
<td>411 keV</td>
<td>Source</td>
<td>(7.17 ± 0.12)( \times )10(^3)</td>
<td>(1.72 ± 0.03)( \times )10(^3)</td>
<td>(1.96 ± 0.03)( \times )10(^3)</td>
</tr>
<tr>
<td>gas</td>
<td>(6.87 ± 0.01)( \times )10(^4)</td>
<td>(5.47 ± 0.01)( \times )10(^4)</td>
<td>(9.02 ± 0.07)( \times )10(^4)</td>
<td></td>
</tr>
<tr>
<td>675 keV</td>
<td>Source</td>
<td>(6.17 ± 0.33)( \times )10(^3)</td>
<td>(3.17 ± 0.17)( \times )10(^3)</td>
<td>(1.13 ± 0.01)( \times )10(^3)</td>
</tr>
<tr>
<td>gas</td>
<td>(5.18 ± 0.03)( \times )10(^4)</td>
<td>(4.63 ± 0.03)( \times )10(^4)</td>
<td>(6.18 ± 0.12)( \times )10(^4)</td>
<td></td>
</tr>
<tr>
<td>1087 keV</td>
<td>Source</td>
<td>(5.99 ± 0.52)( \times )10(^3)</td>
<td>(3.77 ± 0.33)( \times )10(^3)</td>
<td>(8.20 ± 0.10)( \times )10(^3)</td>
</tr>
<tr>
<td>gas</td>
<td>(4.16 ± 0.08)( \times )10(^4)</td>
<td>(3.84 ± 0.07)( \times )10(^4)</td>
<td>(4.12 ± 0.49)( \times )10(^4)</td>
<td></td>
</tr>
</tbody>
</table>

### Table IV

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Au wire</th>
<th>Al-Au wire</th>
<th>Au foil</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \varepsilon_{\beta\gamma} )</td>
<td>(0.801 ± 0.009)%</td>
<td>(0.242 ± 0.003)%</td>
<td>(2.065 ± 0.029)%</td>
</tr>
<tr>
<td>( \varepsilon_{\gamma\gamma} )</td>
<td>(0.689 ± 0.031)%</td>
<td>(0.383 ± 0.017)%</td>
<td>(1.212 ± 0.011)%</td>
</tr>
<tr>
<td>( \varepsilon_{\gamma\gamma} )</td>
<td>(0.721 ± 0.050)%</td>
<td>(0.495 ± 0.034)%</td>
<td>(0.941 ± 0.015)%</td>
</tr>
</tbody>
</table>

### B. Efficiency for \( \gamma \)-Rays

Since the \( \gamma \)-sensitivity \( \varepsilon_{\gamma\gamma} \) of a \( \beta \)-detector is due to the detection of photoelectron and Compton electrons produced by the \( \gamma \)-ray interactions with surrounding objects, the overall efficiency \( \varepsilon_{\gamma\gamma} \) consists of four main components [7]: \( \varepsilon_{\gamma\beta\gamma} \), \( \varepsilon_{\gamma\gamma\gamma} \), \( \varepsilon_{\gamma\gamma\gamma\gamma} \), and \( \varepsilon_{\gamma\gamma\gamma\gamma\gamma} \) due to the counts produced by the interactions with the source material, source mount, counting gas and counter wall. Urquhart further indicated that the combined effect of wall and gas is a fixed quantity inherent in a particular counter with the same counting gas used and is therefore called intrinsic efficiency \( \varepsilon_{\gamma\beta\gamma} \).

The probabilities of escape photoelectrons and Compton electrons into the counting gas per initial photon are shown in Table III. These probabilities were taken as the efficiency \( \varepsilon_{\gamma\beta\gamma} \) and \( \varepsilon_{\gamma\gamma\gamma} \) respectively. The \( \gamma \)-ray interactions with source mount are extremely small and ignored (\( \varepsilon_{\gamma\gamma\gamma\gamma} \approx 0 \)). \( \varepsilon_{\gamma\gamma\gamma\gamma} \) was taken from Urquhart’s semi-empirical value [7] for the same counter. The values were \( \varepsilon_{\gamma\gamma\gamma\gamma} \approx 0.015\% ± 0.003\% \), \( 0.020\% ± 0.001\% \) and \( 0.080\% ± 0.002\% \) for 411 keV, 675 keV and 1087 keV photons respectively. It can be seen that the \( \gamma \)-ray interactions with the source material is the major component.

\( \varepsilon_{\beta\gamma} \) for the pure Au wire, Al-Au wire and pure Au foil obtained by summing up \( \varepsilon_{\gamma\beta\gamma} \) and \( \varepsilon_{\gamma\gamma\gamma} \), and \( \varepsilon_{\gamma\gamma\gamma\gamma} \) is listed in Table IV. Au foil has the highest \( \gamma \)-sensitivity over the other two types of sources. The advantage of using Monte Carlo to calculate the \( \gamma \)-sensitivity over the conventional absorber foil method [21]–[23] is that it overcomes the problem of photon interaction with the absorbers.

The efficiency ratios \( \frac{\varepsilon_{\gamma\gamma}}{\varepsilon_{\gamma\gamma}} \) and \( \frac{\varepsilon_{\gamma\gamma}}{\varepsilon_{\gamma\gamma}} \) were obtained experimentally [8] and corrected for the source absorption of \( \gamma_1 \), \( \gamma_2 \), and \( \gamma_3 \). The corrected values of \( \varepsilon_{\gamma\gamma}/\varepsilon_{\gamma\gamma} \) and \( \varepsilon_{\gamma\gamma}/\varepsilon_{\gamma\gamma} \) were 0.16 and 0.09 respectively for all materials. The uncertainties in \( \varepsilon_{\gamma\gamma}/\varepsilon_{\gamma\gamma} \) and \( \varepsilon_{\gamma\gamma}/\varepsilon_{\gamma\gamma} \) have minor influence on the
activity value. For example, an uncertainty of 20% contributes less than 0.02% to the total uncertainty of the activity value.

C. Activity Determination

The nuclear decay data used for the correction term are taken from [1].

Using the values discussed before, \( g, f, h, \) and \( K \) were determined and listed in Table V. The corrections for pure Au wire and foil are both high. However they are due to different mechanism. Au wire has high absorption of beta particles, whereas Au foil gives high \( \gamma \)-sensitivity of the \( \beta \)-detector. The correction \( K \) is strongly influenced by the beta efficiency \( \varepsilon_{\beta\alpha} \) of the major decay branch (see Fig. 5).

The activity for each sample was determined using (4) with \( N_{\beta}, N_{\gamma}, \) and \( N_{c} \) obtained experimentally from the 4\( \pi \beta-\gamma \) coincidence counting measurements. \( N_{\beta}, N_{\gamma}, \) and \( N_{c} \) were corrected for dead time, background and accidental coincidences.

Several photon emitting impurities were detected in the Al-Au samples using the HPGe gamma-ray spectroscopy (see Fig. 6). They were 0.2% of \( ^{60} \)Co, 0.5% of \( ^{182} \)Ta, 0.4% of \( ^{199} \)Au and 0.1% of \( ^{181} \)Hf at the 4\( \pi \beta-\gamma \) coincidence measurement reference time. A total correction of 1,0033 ± 10% for these impurities was applied. Of these impurities, \( ^{199} \)Au was produced through the reaction \( ^{198} \)Au (n, \( \gamma \))\(^{199} \)Au as \( ^{198} \)Au has a very large thermal neutron capture cross section (2,6 × 10^4 barn [24]). Pure Au samples have negligible amount of \( ^{199} \)Au due to short irradiation time. Other impurities were produced through the neutron activation of the chemical impurities in the Al-Au alloy.

The uncertainties were evaluated in accordance to the Guide to the Expression of Uncertainty in Measurement [25].

According to the MCNP calculations, for the wires, the uncertainty of probability of escape electrons due to deviation in diameter is significant, whereas in length is minimal. However, for the foils, uncertainties due to both dimensions are of similar significance.

The major uncertainty components for \( N_{\beta}, N_{\gamma}, \) and \( N_{c} \) are counting statistics (0.6%), dead time (0.5%), resolving time (0.8%) and half life (0.1%). These give a combined relative uncertainty of 0.7%, 0.6% and 0.7% in \( N_{\beta}, N_{\gamma}, \) and \( N_{c} \) respectively. Combining the uncertainties of \( N_{\beta}, N_{\gamma}, \) and \( N_{c} \), and \( K \) gives the total relative uncertainty of the activity of 0.7%, 0.5%, and 0.8% for Au wire, Al-Au wire and Au foil respectively.

For comparison, Table VI lists the value of \( \varepsilon_{\beta\alpha}, \varepsilon_{\beta\gamma}, \) and \( K \) for the foils from this work and publications [5], [6], [8]. We note that the efficiency from these publications, obtained from equation \( \varepsilon_{\beta\alpha} = N_{c}/N_{\gamma} \), were not corrected. We highlight that, in this work, the high correction \( K \) value arises from the high \( \gamma \)-sensitivity of the \( \beta \)-detector calculated using MCNP.

D. Comparison With the Experimental Results

The experimental method, namely the 4\( \pi \beta-\gamma \) coincidence counting efficiency extrapolation method, was used to measure the absolute activity of pure Au wire samples by dissolving the wires with a aqua-regia solution [26]. The standard developed in this way was registered in the ANSTO Secondary Standard TPA ionization chamber as a calibration factor (in the unit of pA/MBq) for pure Au wires.

The activities for the three Au wire samples determined using the method described in this work were compared with the activities determined by the ionization chamber measurements. Both
activity values were agreed with each other better than 0.2%, indicating the correctness of the simulation model used in this work.

IV. CONCLUSION

Absolute activities of solid sources are difficult to measure due to the source self-absorption and γ-ray interaction with the source materials. This work has demonstrated that the absolute activities of solid sources, such as $^{108}$Au in the form of Au wire, Al-Au alloy wire and Au foil, can be determined using irradiation of the samples, and Mr. M. Smith for assisting in the porting this work, Dr. G. Braoudakis for providing guidance in work.

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REFERENCES


[10] H. Miyahara, M. Mizuno, and T. Watanabe, “Calculation of efficiency functions in $4\pi\beta$—$\gamma$ coincidence counting method with the correction term of the coincidence equation being calculated by Monte Carlo simulation technique. The Monte Carlo simulation of the absorption of electrons and gamma photon interactions with the surrounding materials allows the efficiencies for $\beta$-particles, conversion electrons and the $\gamma$-sensitivity of the $\beta$-detector to be calculated. The efficiency of the main branch $\beta$-particles can be obtained experimentally with a small correction determined using the Monte Carlo calculations and its uncertainty is the dominant component for the correction term.

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Chapter Six

Characterising neutron flux for the hot commissioning of the OPAL reactor

L. Mo, M. Smith, L. Bignell, M. J. Qin, D. Alexiev

Australian Nuclear Science and Technology Organisation, New Illawarra Road, Lucas Heights, NSW 2234, Australia

Author’s contribution

L. Mo (candidate)
Contributed to the experimental design and carried out experiment, analysed experimental data and wrote manuscript.

M. Smith
Contributed to the experimental design, carried out experiment and data analysis.

L. Bignell
Carried out experiment and data analysis.

M.J. Qin
Contributed to experiment and data analysis.

D. Alexiev
Contributed to experimental design and carried out experiment.
Characterising Neutron flux for the hot commissioning of the OPAL reactor

L. Mo
M. Smith
L. Bignell
M. J. Qin
D. Alexiev

Radiation Physics

SAFETY AND RADIATION SERVICES

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Characterising Neutron flux for the hot commissioning of the OPAL reactor

ABSTRACT

For the commissioning process of the new nuclear reactor, OPAL (Open Pool Australian Light-water) nuclear reactor, at the Australian Nuclear Science and Technology Organisation (ANSTO), the thermal neutron flux and fast neutron flux are determined by the activity measurement of activated Al-Au (0.112% of Au) alloy wires and Ni wires.

The neutron flux $\phi_{0.625}$ (for neutron energy 0 – 0.6 eV) distribution, epithermal neutron flux distribution and fast neutron flux distribution were determined for the ‘startup’ core of the OPAL reactor and irradiation facilities. The power peaking factor (PPF) of the “startup” core is 1.9, which meets the reactor design. The flux distribution in the irradiation facilities has shown the quality of the reactor design.
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1. Introduction

The OPAL (Open Pool Australian Light-water) reactor is a 20 MW, open-pool research reactor. It is used for radioisotope production, irradiation services and neutron beam research. The reactor uses low enriched uranium fuel in a compact core, cooled with light water and moderated by heavy water, yielding maximum thermal neutron flux of more than $10^{14}$ n/cm$^2$s [Kim 2006].

As shown in Figure 1, the reactor core consists of 16 fuel elements, each made of 21 low-enriched uranium silicide ($\text{U}_3\text{Si}_2$) fuel plates with aluminium cladding. The active dimensions of each fuel plate are 65 mm x 615 mm. Between each plate there are water channels for cooling and moderation. The core measures 350 x 350 x 615 mm and is located inside the reactor pool at 10 m below water surface level. Reactivity is controlled by five independent control blades driven from underneath, four of which have hafnium plates, and the fifth has a central cruciform shaped plate.

![Figure 1. OPAL core configuration with indication of high power density and high thermal neutron flux [INVAP, 2005].](image)

In the reactor, there are 6 NTD (neutron transmutation doping) Si irradiation facilities, 17 bulk irradiation facilities (BIF), 2 positions for neutron activation analysis and 55 low flux...
facilities (LF) for general purposes. These irradiation facilities are located in the heavy water reflector vessel.

This paper presents the neutron flux characteristics in the reactor core and irradiation facilities, determined by the activity measurement of the neutron activated Al-Au (0.1124% Au) alloy wires and Ni wire.

2. Method

Thermal and epithermal neutron flux measurements were performed by neutron activation of Al-Au alloy wires (Thermal Electron, 0.1124% Au, diameter 0.508 mm and length 7.7 mm) with and without cadmium covers. Cadmium was used to absorb thermal neutrons. Fast neutron flux measurements were performed by neutron activation of Ni wires (Thermo Electron, 99.993% Ni, diameter 0.762 mm and length of 8.0 mm). Some pure Au wires (Thermo Electron, 99.9971% Au, diameter 0.125 mm and length 8.0 mm) were used for neutron detector calibration (2.3 W) at the reactor core. The Al-Au and Ni wires are called flux monitors.

2.1. Sample loading in the “startup” core

A neutron flux distribution was measured along the vertical dimension of the fuel plates, where the possible power density is above 90% of the highest power density. According to the calculations made by the INVAP [INVAP, 2005], these fuel plates are in the fuel elements A2, B4 and D2 (see Figure 1). The flux monitors (Al-Au wires) were placed in the channel 20 of elements A2, B4 and D2, the channel nearest to these fuel plates. A neutron flux distribution was also measured along the vertical dimension of the fuel plates, where the possible thermal neutron flux is above 90% of the highest thermal neutron flux. According to the calculations, these fuel plates are in element B1 and C1 (see Figure 1). The flux monitors (Al-Au wires) were placed in channel 1 of B1 and C1, the channel nearest to these fuel plates. The positions of the peak (highest) power density in channel 20 of element A2, B4 and D2 are expected either in 18 cm or 9 cm below the vertical core centre. A flux distribution in the horizontal dimension of these positions was measured. A plane mapping was undertaken by measuring the neutron flux in 9 cm and 18 cm below vertical core centre in channel 8 or 13 of each element.

Approximately 150 Al-Au wires were placed on 16 aluminium plates using sticky tape. Of these wires 51 were covered with cadmium tubes for the determination of epithermal neutron flux and cadmium ratio and 16 bare wires were used to determine the normalisation factor for two different irradiations. The epithermal neutron flux is measured in the axial position of 24
cm, 12 cm, 0 cm, -12 cm, -24 cm with reference to the core centre. The epithermal neutron flux in other positions was obtained by linear interpolation or extrapolation of these values.

The aluminium plates loaded with Al-Au were inserted into certain water channel of each fuel element. Two irradiations were undertaken; one for bare Al-Au wires and one for cadmium covered Al-Au wires. The cadmium covered wires are spread over the two irradiations in order to reduce the flux and reactivity perturbation due to the cadmium tubes. The wires were irradiated at approximately 40 kW for 2 hours.

2.2. Sample loading in the irradiation facilities

The effective length of the NTD Si irradiation facilities is 600 mm. As demonstrated in Figure 2, for the vertical neutron flux profile in the NTD Si irradiation facilities, some 40 Au-Al wires were placed along the axial direction on the surface of dummy Si ingots with 2 cm apart from each other. Two columns with $120^\circ$ away for column B from A were mapped. 11 cadmium covered Al-Au wires were placed at 6 cm apart from each other in the column C for the measurements of epithermal neutrons. 10 Ni wires covered with cadmium were also placed in the column C between cadmium covered Al-Au wires for the measurement of the fast neutron flux. The radial flux profiles were measured at several axial positions and the wires were placed on the radial surface of the ingot along the diameter.

The wires were irradiated for 2 hours at power of 20 MW. The ingots were rotated during irradiation time.

![Diagram](image)

Figure 2. Illustration of neutron flux profiling position in NTD Si irradiation facilities.
For other irradiation facilities, the axial and radial flux profiles were measured using Al-Au wires. Figure 3 illustrates the radial positions where the Al-Au wires were allocated.

![Diagram of sample loading for radial neutron flux distribution of BIF and LF irradiation facilities.](image)

**Figure 3. Illustration of sample loading for radial neutron flux distribution of BIF and LF irradiation facilities.**

### 2.3. Activity measurements of $^{198}$Au

Activity measurements of the activated Al-Au and Ni wires were carried out in two GEM Series HPGe coaxial crystal detectors (Model GEM-15200 and GEM-18180-P). The distance between source and detector end cap is 100 mm.

The HPGe detectors were calibrated for 411 keV full energy photopeak using the standardised sources, which had exactly the same geometry as the measured samples. The standardisation of the calibration sources in the form of pure Au wire and Al-Au alloy wire were performed using $4\pi\beta-\gamma$ coincidence counting technique with the correction term determined by the Monte Carlo simulation technique [Mo et al., 2007]. The direct calibration of the HPGe detectors against the sources with the same geometry eliminates the need of correction for efficiency loss due to coincidence summing effects.

The fast neutron flux measurements did not require high accuracy. The detection efficiency for 810 keV full energy photopeak (for the activity measurement of $^{58}$Co) was obtained by interpolation of the efficiency function. The efficiency function was obtained using a set of standard point sources from Oak Ridge (Tennessee, USA). The distance between source and detector end cap was 100 mm.

The efficiencies for the four detectors are listed in table 1.

<table>
<thead>
<tr>
<th>Detector</th>
<th>411 keV efficiency</th>
<th>810 keV efficiency</th>
</tr>
</thead>
<tbody>
<tr>
<td>GEM-15200</td>
<td>0.25%</td>
<td>0.11%</td>
</tr>
<tr>
<td>GEM-18180-P</td>
<td>0.28%</td>
<td>0.13%</td>
</tr>
</tbody>
</table>
2.4. Neutron flux calculations

The thermal neutron flux was measured by gold activation according to the ASTM standard [ASTM International, 2006a]. The reaction is represented by $^{197}\text{Au} (n, \gamma) ^{198}\text{Au}$.

For a neutron monitor irradiated by neutrons for a period of time $t_{i}$ and counted in a HPGe gamma-ray spectrometer after waiting time $t_{w}$ from the end of irradiation, the reaction rate $R$ per analysed element atom is given by

$$R = \frac{MAe^{\lambda_{w}}}{N_{A}mc(1 - e^{-\lambda_{i}})},$$  
(1)

where,
- $M$: Molar mass of the specific stable isotope in the neutron detector;
- $\lambda$: decay constant of the produced radioactive isotope;
- $N_{A}$: Avogadro number ($6.022045 \times 10^{23}$ Atoms/mol);
- $m$: total mass of the neutron detector;
- $c$: concentration of the specific stable isotope in the neutron detector;
- $t_{i}$: irradiation time;
- $t_{w}$: waiting time between the end of irradiation and the start of the activity measurements;
- $A$: activity of the produced isotope at the measurement time.

For the reaction rate $R_{s,c,d}$ and $R_{s}$ obtained by irradiating gold detector with and without cadmium cover and calculated using formula (1), the conventional thermal neutron flux $\phi_{0}$ and the epithermal neutron flux $\phi_{e}$ are given by

$$\phi_{0} = \frac{1}{G_{th}G\sigma_{0}} \left[ R_{s} - R_{s,c,d} \left( 1 + \frac{g\sigma_{0}}{G_{res}I_{0}} f_{1} + \frac{\sigma_{0}W'}{G_{res}I_{0}} \right) \right],$$  
(2)

and

$$\phi_{e} = \frac{R_{s,c,d}}{G_{res}I_{0}},$$  
(3)

where,
- $g$: the correction factor for the departures from the ideal $1/\nu$ detector cross section in the thermal energy range. $g = 1.0051$. 


\( \sigma_0 \): the 2200 m/s cross section (as normally tabulated in neutron cross section data);

\( \phi_e \): epithermal flux parameter;

\( f_1 \): a function that describes the epithermal activation of a 1/\( v \) detector in the energy range \( 5kT \) to \( E_{cd} \) (cadmium cut-off energy),

\[
\phi_e = \phi_{e0} \left( \frac{kT}{E} \right)^{1/2} \frac{dE}{E}; \\
\text{For } E_{cd} = 0.43 \text{ eV, } T_0 = 293.4^\circ \text{K and } 5kT = 0.13 \text{ eV, } f_1 = 0.397;
\]

\( w' \): a function which accounts for the departure of the cross section from the 1/\( v \) law in the energy range \( 5kT \) to \( E_{cd} \),

\[
w' = \frac{1}{\sigma_0} \int_{5kT}^{E_{cd}} \sigma(E) - \sigma_0 \left( \frac{kT}{E} \right)^{1/2} \frac{dE}{E}; \\
\text{For } E_{cd} = 0.43 \text{ eV, } T = 293.4^\circ \text{K and } 5kT = 0.13 \text{ eV, } w' = 0.032;
\]

\( I_0 \): the resonance integral,

\[
I_0 = \int_{E_{cd}}^{\infty} \sigma(E) \frac{dE}{E} = 1557 \pm 30 \text{ barn};
\]

\( G_{th} \) and \( G_{res} \): thermal self-shielding and epithermal self-shielding factors respectively (1 and 1 for Al-Au wire, and 0.95 and 0.278 for pure Au wire respectively).

In a well-thermalized reactor system, the neutron flux spectrum for neutron temperature \( T \) can be assumed to be the sum of a Maxwellian distribution in the neutron energy range \( 0 < E < \infty \) and Epithermal spectrum in the range \( 5kT < E < \infty \), which may be expressed by [Beckurts and Wirtz, 1964]

\[
\phi(E) = 1.128 \sqrt{\frac{T}{T_0}} \phi_0 \frac{E}{(kT)^2} e^{-E/kT} + \phi_e \frac{\Delta(E/kT)}{E}, \tag{4}
\]

where, \( k \) is the Boltzmann’s constant and \( \Delta(E/kT) \) is called the “joining” function. \( \Delta(E/kT) = 0 \) for \( E < 5kT \) and \( \Delta(E/kT) = 1 \) for \( E > 5kT \) [INVAP, 2004].

The neutron flux for the energy \( 0 - 0.625 \) eV is used for the assessment of the reactor design. It is obtained by integration of the neutron flux spectrum (equation (4)) over energy range \( 0 \) to \( 0.625 \) eV and given by

\[
\phi_{0.625} = 1.128 \sqrt{\frac{T}{T_0}} \phi_0 + 1.5702 \phi_e. \tag{5}
\]

The fast neutron flux was measured by nickel activation according to the ASTM E264-02 Standard [ASTM International, 2006b]. The reaction is represented by \(^{58}\text{Ni} (n, p)^{58}\text{Co} \). The flux value is given by
\[
\phi_f = \frac{R}{\sigma}, \quad (6)
\]
where, \( R \) is the reaction rate obtained from equation (1) and \( \sigma = 0.113 \pm 0.003 \) barn [Baard et al., 1992] which is the mean cross section for the nickel reaction.

3. Results and discussion

The count rates measured in the gamma detectors were corrected for system dead time and radionuclide decay during the measurement period using the formulae recommended by Wehe et al. [Wehe et al., 1992]. The activities were in the level of \( 10^4 - 10^5 \) Bq for the bare Al-Au wires and Au foils, \( 10^3 - 10^4 \) Bq for the cadmium covered Al-Au wires and \( 10^2 \) Bq for Ni wires. The uncertainties of the measured activities are approximately 1.4%, contributed dominantly from the uncertainties in the detector efficiency and counting statistics.

Based on these activities, the following parameters were determined: 1) the reaction rate using equation (1); 2) conventional thermal neutron flux \( \phi_0 \) using equation (2); 3) epithermal neutron flux \( \phi_e \) using equation (3); 4) cadmium ratio \( \frac{R_{cd}}{R_{s,cd}} \); 4) neutron flux \( \phi_{0.625} \) for the neutron energy \( 0 - 0.625 \) eV using equation (5). The standard uncertainty for each value was evaluated according to the ISO ‘Guide to the Expression of Uncertainty in Measurement’ [International Organisation for Standardisation, 1993].

3.1. Neutron flux distribution in the “startup” core

Figure 4 shows the neutron flux \( \phi_{0.625} \) distribution and epithermal neutron flux \( \phi_e \) distribution along the vertical dimension of the fuel elements 1) A2, water channel 20; 2) C1, water channel 1; 3) B1, water channel 1; 4) B4, water channel 20; and 5) D2, water channel 20. Figure 5 shows the neutron flux \( \phi_{0.625} \) and \( \phi_e \) distribution in the horizontal dimension of the water channel at the position of 18 cm below the vertical core centre for the fuel elements 1) A2, channel 20; 2) D2, channel 20; and 9 cm below the centre for the element 3) B4, channel 20.
Figure 4. (a) Neutron (0 - 0.625 keV) and (b) epithermal neutron flux distribution along vertical dimension of the core for the fuel element B1, C1, B4, A2 and D2.
From Figure 4 and 5, we can see that B1 and C1 have similar neutron flux profile, and A2 and D2 have similar neutron flux profile. The maximum neutron flux $\phi_{0.625}$ and $\phi_e$ is $3.0 \times 10^{14}$ n/cm$^2$/s and $2.1 \times 10^{13}$ n/cm$^2$/s respectively at 12 cm below the vertical core centre in fuel element B1 and C1. The minimum is $4.0 \times 10^{13}$ n/cm$^2$/s and $4.5 \times 10^{12}$ n/cm$^2$/s respectively at 30 cm above the vertical core centre in the element D2. The flux value $\phi_{0.625}$ and $\phi_e$ obtained in the core plan mapping in 18 and 9 cm below the vertical core centre in each fuel element are in the range of
9.9\times10^{13} \text{ to } 1.89\times10^{14} \text{ and } 1.5\times10^{13} \text{ to } 2.3\times10^{13} \text{ n/cm}^2\text{s respectively. All neutron flux values were measured at power level of 40 kW and scaled up to 20 MW full reactor power. The cadmium ratio is between 1.2 and 2.0 for the whole core mapping. The power peaking factor (PPF, the ratio of maximum power over average power of the core) of the “startup” core is 1.9, which meets the reactor design.

The uncertainty of $\phi_{0.625}$ was approximately in the range of 3.2% - 8.4% depending on the cadmium ratio $R_{cd}$ when other experimental conditions are the same. Cadmium ratio influences the overall uncertainty of the neutron flux through the sensitivity coefficient of the reaction rate. When $R_{cd} > 100$, the uncertainty of $\phi_{0.625}$ is independent of $R_{cd}$. The predominant sources of uncertainty in $\phi_{0.625}$ are the uncertainty of 411 keV full-energy peak efficiency of HPGe detector, net counts and power ramp. The uncertainty of epithermal neutron flux $\phi_e$ is 2.4%. The full uncertainty analysis of true thermal neutron flux, epithermal neutron flux and neutron flux $\phi_{0.625}$ is described by Mo et al. [Mo et al, 2006].

3.2. Neutron flux distribution in the irradiation facilities

The neutron flux measurements were carried out in all 80 irradiation facilities. Figure 6 shows the vertical and radial thermal neutron flux $\phi_{0.625}$ distribution in one of the NTD Si irradiation facilities, NTD-2. The flux was measured by placing the Al-Au wires on the cylindrical and radial surface of the silicon ingot. The flux is normalised to the average flux of the facility. It can be seen that the rotation of the Si ingot made the flux received by the two columns similar. The maximum deviation from the average flux for each column is $\pm$ 5%, which agrees with the contractual design (5%).

![Figure 6. Normalised axial neutron flux ($\phi_{0.625}$) profile of NTD-2 irradiation facility.](image-url)
The fast neutron flux distribution in NTD-2 is shown in Figure 7. The ratio of thermal neutron flux ($\phi_{0.625}$) to the fast neutron flux in the same position is greater than 800 which ensures minimum damage to the NTD Si products.

![Figure 7. Axial fast neutron flux distribution of NTD-2 irradiation facility.](image)

Figure 8 shows the neutron flux $\phi_{0.625}$ distribution along the axis and in the radial plane for one of the low flux (LF) irradiation facilities, LF11. The flux distribution meets the contractual design.

![Figure 8. Neutron flux distribution along the axis and in the radial plane.](image)
4. Conclusion

The neutron flux $\phi_{0.625}$ (for the neutron energy $0 – 0.625$ eV) distribution, epithermal neutron flux distribution and fast neutron distribution was determined for the ‘start-up’ core of the OPAL reactor and irradiation facilities. The thermal neutron flux was measured by neutron activation of Al-Au alloy wires with and without cadmium covers. The fast neutron flux was measured by neutron activation of Ni wires with cadmium covers. The power peaking factor (PPF) of the “startup” core is 1.9, which meets the reactor design. The flux distributions in the irradiation facilities have also shown the quality of the reactor design.

Reference


Mo, L., Alexiev, D., Baldock, C., 2006. Uncertainty evaluation of thermal neutron flux determination over the energy range 0 – 0.6 eV. International Metrology conference, 3 – 7 April, 2006, Casablanca.


Chapter Seven

Uncertainty evaluation of thermal neutron flux determination over the energy range 0 – 0.625 eV

L. Mo\textsuperscript{a, b}, D. Alexiev\textsuperscript{a}, C. Balock\textsuperscript{b}

\textsuperscript{a}Australian Nuclear Science and Technology Organisation, New Illawarra Road, Lucas Heights, NSW 2234, Australia

\textsuperscript{b}Institute of Medical Physics, School of Physics, University of Sydney, NSW 2006, Australia

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Author’s contribution

L. Mo (candidate)
Analysed theoretical formulas and data, wrote manuscript.

D. Alexiev
Contributed to manuscript editing.

C. Baldock
Edited manuscript.
Uncertainty evaluation of thermal neutron flux determination over the energy range 0 – 0.625 eV

L. Mo\textsuperscript{a,b}, D. Alexiev\textsuperscript{a}, C. Baldock\textsuperscript{b}

\textsuperscript{a}Australian Nuclear Science and Technology Organisation, New Illawarra Road, Lucas Heights, NSW 2234, Australia
\textsuperscript{b}Institute of Medical Physics, School of Physics, University of Sydney, NSW 2006, Australia

Abstract

At the commissioning stage of the OPAL nuclear reactor of the Australian Nuclear Science and Technology Organisation (ANSTO) in 2006, the thermal neutron flux distribution will be assessed through the activity measurement of activated Au-Al alloy wires with Au concentration of 0.112%. The activity will be determined using a HPGe gamma ray spectrometer. Being equally important as the flux value itself, the uncertainty of the thermal neutron flux over the energy range 0 to 0.6 eV is analysed. The predominant uncertainty components are the uncertainty of the 411 keV full-energy peak efficiency, net counts and the uncertainty due to power ramps. Reducing these uncertainties will improve the accuracy of the thermal neutron flux determination.

Keywords: uncertainty, reaction rate, neutron flux, activity measurement,

1. Introduction

The commissioning of the OPAL nuclear reactor at the Australian Nuclear Science and Technology (ANSTO) will be taking place in 2006. Neutron flux distribution is a very important characteristic to validate the accuracy of the reactor design and verify the research performance as a neutron source. At the commissioning stage, the neutron flux distribution in the core and irradiation facilities will be assessed through the activity measurement of activated Au-Al alloy wires with Au concentration of 0.112%.

There will be hundreds of Au-Al alloy wires to be irradiated and measured in a short period. The time constraint requires optimum selection of neutron irradiation and activity measurement parameters. This paper describes the sources of uncertainty of thermal neutron flux and the influence of the uncertainty components to the overall uncertainty in order to provide the directions of effectively improving the accuracy of neutron flux determination.

2. Neutron flux determination

Gold activation involves capture of the neutron by \(^{197}\text{Au}\) and production of \(^{198}\text{Au}\), followed by the emission of gamma radiation through the decay of \(^{198}\text{Au}\) into \(^{198}\text{Hg}\). The reactions are represented by \(^{197}\text{Au}\,(n, \gamma)\) \(^{198}\text{Au}\) for nuclear reaction and \(^{198}\text{Au} \rightarrow ^{198}\text{Hg} + \beta^- + \gamma\) for disintegration.

In a well-thermalized reactor system, the neutron flux spectrum for neutron temperature \(T\) can be assumed to be the sum of a Maxwellian distribution in the neutron energy range \(0 < E < \infty\) and Epithermal spectrum in the range \(5kT < E < \infty\), which may be expressed by a function [1] of

\[
\phi(E) = \phi_h \frac{E}{(kT)^2} e^{-E/kT} + \phi_e \frac{\Delta(E/kT)}{E},
\]

where, \(\phi_h\) is the Maxwellian thermal neutron flux, \(\phi_e\) the epithermal neutron flux, \(T\) the neutron temperature, \(k\) the Boltzmann’s constant and \(\Delta(E/kT)\) is called the “joining” function. \(\Delta(E/kT) = 0\) for \(E < 5kT\) and \(\Delta(E/kT) = 1\) for \(E > 5kT\) [2]

With the conventional neutron flux defined [3] by the expression \(\phi_0 = n_0\nu_0\), namely the product of the Maxwellian thermal neutron density \(n_0\) and the most probable neutron speed \(\nu_0\) (2200 m/s) for the Maxwellian thermal neutron distribution at the ordinary neutron temperature \(T_0 = 293.4^0K\), the Maxwellian thermal neutron flux at a temperature \(T\) is related to the conventional neutron flux in the form [4] of

\[
\phi_h = \frac{2}{\sqrt{\pi}} \frac{T}{T_0} \phi_0 = 1.128 \frac{T}{T_0} \phi_0,
\]
where, \(2/\sqrt{\pi}\) is the ratio of the average velocity in a Maxwellian distribution to the most probable velocity. The neutron flux spectrum at temperature \(T\) then becomes

\[
\phi(E) = 1.128 \frac{\int T \phi_0 E}{(kT)^{3/2}} e^{-E/kT} + \phi_e \frac{\Delta(E/kT)}{E}.
\]  

The reaction rate per atom, for an isotope exposed to a mixed thermal and epithermal neutron field, is given by [3]

\[
R_e = \phi_0 g \sigma_0 + \phi_e g \sigma_0 \left( f_1 + \frac{w'}{g} \frac{l_0}{\sigma_0} \right),
\]  

where,

- \(g\): the correction factor for the departures from the ideal \(1/v\) detector cross section in the thermal energy range, depending on the neutron temperature \(T\), is given by

\[
g = \frac{1}{\nu_0 \sigma_0} \left[ \frac{\pi}{4} \frac{\nu}{\nu_0} \left( \frac{T_0}{T} \right)^{3/2} \right] \exp \left[ -\left( \frac{\nu}{\nu_0} \right)^2 \left( \frac{T_0}{T} \right) \right] \sigma(v) dv;
\]

- \(\sigma_0\): the 2200 m/s cross section (as normally tabulated in neutron cross section data);

- \(\phi_e\): epithermal flux parameter;

- \(f_1\): a function that describes the epithermal activation of a \(1/v\) detector in the energy range \(5kT\) to \(E_{cd}\) (cadmium cut-off energy),

\[
f_1 = \frac{E_{cd}}{5kT} \left( \frac{kT_0}{E} \right)^{1/2} \frac{dE}{E} \; \text{for} \; E_{cd} = 0.43 \; \text{eV}, \; T_0 = 293.4^\circ \text{K} \; \text{and} \; 5kT = 0.13 \; \text{eV}, \; f_1 = 0.397;
\]

- \(w'\): a function which accounts for the departure of the cross section from the \(1/v\) law in the energy range \(5kT\) to \(E_{cd}\),

\[
w' = \frac{1}{\sigma_0 5kT} \int_{E_{cd}}^{\infty} \sigma(E) - g \sigma_0 \left( \frac{kT}{E} \right)^{1/2} \frac{dE}{E}; \; \text{for} \; E_{cd} = 0.43 \; \text{eV}, \; T = 293.4^\circ \text{K} \; \text{and} \; 5kT = 0.13 \; \text{eV}, \; w' = 0.032;
\]

- \(l_0\): the resonance integral,

\[
l_0 = \int_{E_{cd}}^{\infty} \sigma(E) \frac{dE}{E} = 1557 \pm 30 \; \text{barn};
\]

For a Au-Al alloy wire irradiated by neutrons for a period of time \(t_i\) and counted in a HPGe gamma-ray spectrometer after waiting time \(t_w\) of the end of irradiation, the reaction rate \(R\) per analysed element atom is given by

\[
R = \frac{M_0 e^{\lambda t_w}}{N_A mc(1 - e^{-\lambda t_i})},
\]  

where,

- \(M\): Molar mass of \(^{197}\text{Au}\) (196.9665 ± 0.0001 g/Mol);

- \(\lambda\): decay constant of \(^{198}\text{Au}\) (1.7865E-04 ± 0.0005E-04 min);

- \(N_{A}\): Avogadro number (6.022045x10\(^{23}\) Atoms/mol);

- \(m\): mass of the target;

- \(c\): concentration of the Au in the target (0.112%);

- \(t_i\): irradiation time;

- \(t_w\): waiting time between the end of irradiation and the start of the activity measurements;

- \(A\): activity of the activated Au-Al wire at the measurement time, is given by \(A = C \epsilon \phi_t\), where, \(C\) denotes the net counts under the \(^{198}\text{Au}\) 411 keV photopeak measured with a HPGe \(\gamma\)-ray spectrometer, \(\phi_t\) is the system live time, \(p\) is 411 keV photon emission probability and \(\epsilon\) is the 411 keV full-energy peak efficiency.

For the reaction rate \(R_{cd}\) and \(R_e\) obtained by irradiating Au-Al alloy wires with and without cadmium cover and calculated using formula (6), the conventional neutron flux \(\phi_0\) and the epithermal neutron flux \(\phi_e\) are given by

\[
\phi_0 = \frac{1}{G_{th} \sigma_0} \left[ R_e - R_{cd} \left( 1 + \frac{g \sigma_0}{G_{res} l_0} f_1 + \frac{\sigma_0 w'}{G_{res} l_0} \right) \right],
\]  

and

\[
\phi_e = \frac{R_{cd}}{G_{res} l_0},
\]  

where \(G_{th}\) and \(G_{res}\) are thermal self-shielding and epithermal self-shielding factors respectively.

Finally, the thermal neutron flux needed to be determined is the flux with neutron energy lower than 0.625 eV. The flux is obtained by the integration of the neutron flux spectrum expressed by equation (3):

\[
\phi_{0.625} = \int_{0}^{0.625} \left[ \frac{T}{T_0} \phi_0 \left( \frac{E}{E_{th}} \right)^{1/2} e^{-E/kT} + \phi_e \frac{\Delta(E/kT)}{E} \right] \frac{dE}{E}
\]

\[
= 1.128 \frac{\int T}{T_0} \phi_0 + 1.570 \phi_e.
\]  

Table 1 lists the physical constants and their uncertainties, mentioned above, for the neutron flux determination. These uncertainties are B type uncertainties and will be used to evaluate the standard uncertainty of \(\phi_{0.625}\).
The evaluation of uncertainty of the non-linear expression of measurand and inputs, the strongly “depends on each of the input variables. The evaluation of uncertainty of reaction rate, in equation (6) and is given by

$$U_R^2 = \left(\frac{U_c}{c}\right)^2 + \left(\frac{U_m}{m}\right)^2 + \left(\frac{U_c}{c}\right)^2 + \left(\frac{U_m}{m}\right)^2 + \left(\frac{U_t}{t}\right)^2.$$

where $U_{A_1}$, $U_{A_2}$, ..., $U_{A_n}$ are the standard uncertainties of $A_1$, $A_2$, ..., $A_n$ and $U(x_i, x_j)$ is the covariance between $x_i$ and $x_j$. The partial derivatives are called ‘sensitivity coefficients’ which show how strongly “y” depends on each of the input variables. The evaluation of uncertainty of $y$ becomes cumbersome if the expression (10) is non-linear. For the non-linear expression of measurand and inputs, the approach adopted in this paper is to evaluate the uncertainty of $y$ to the first order of the Taylor’s expansion of $y$ using function (11).

### 3.1 Uncertainty of reaction rate

In order to evaluate the uncertainty of reaction rate, the uncertainty of activity (A) has to be determined first. The formula for activity determination using a gamma ray detector is $A=C\cdot \rho \cdot \Gamma_t$. Following the ISO Guide the standard uncertainty ($U_A$) of A is then given by

$$\left(\frac{U_A}{A}\right)^2 = \left(\frac{U_C}{C}\right)^2 + \left(\frac{U_{\rho}}{\rho}\right)^2 + \left(\frac{U_{\Gamma}}{\Gamma}\right)^2 + \left(\frac{U_{t}}{t}\right)^2.$$

The photon emission probability term is negligible. The uncertainty of $t_2$ is estimated to be 0.2% and the uncertainty of the detection efficiency is 1.5%, estimation of which is based on the calibration using a $^{198}$Au standard in the form of a pure Au wire. Such a standard was developed by dissolving a pure Au wire and standardising by the $4\pi\beta-\gamma$ coincidence counting technique [7].

With the assumption of Poisson distribution of the disintegration, the standard deviation of the measured net counts about the “true” counts for a single measurement may be expressed by

$$\sigma_c = \sqrt{C}.$$  \hspace{1cm} (13)

For $n$ repeated counting measurements which have similar net counts $C$, the experimental standard deviation of the mean (ESDM) of the net counts is given by

$$u(\bar{X}) = \sqrt{\frac{1}{n} \cdot \sum_{i=1}^{n} \sigma_i^2} \approx \frac{C}{\sqrt{n}}.$$  \hspace{1cm} (14)

For $n = 2$ and $C = 10000$, the relative uncertainty of net counts is 0.7%. Using equation (12), the relative uncertainty of activity is found to be 1.7%.

The standard uncertainty of the reaction rate $R$ per gold atom calculated from the counting data is determined by propagation of the variance of each input in the equation (6) and is given by

$$R = \frac{C}{m} \cdot \rho \cdot \Gamma_t.$$

$$u(R) = \sqrt{\left(\frac{U_c}{C}\right)^2 + \left(\frac{U_{m}}{m}\right)^2 + \left(\frac{U_{\rho}}{\rho}\right)^2 + \left(\frac{U_{\Gamma}}{\Gamma}\right)^2 + \left(\frac{U_{t}}{t}\right)^2 + \left(\frac{U_{w}}{w}\right)^2 + \left(\frac{U_{l}}{l}\right)^2}.$$  \hspace{1cm} (15)

The waiting time and decay constant term in equation (15) are negligible. With the estimation of 0.3% uncertainty in the mass of target $(m)$, 0.3% uncertainty in Au concentration in the target $(c)$ and 0.3% uncertainty in irradiation time $(t_1 = 100$ min), the relative uncertainty of reaction rate is found to be 1.8%.

There are power ramps during the reactor power start up to full power after sample loading and shutdown for sample removal. These ramps introduce additional activity. For a 100 min irradiation, the correction on the reaction rate for the power ramps is 1.4 %, calculated using the formula described in reference [8]. If this correction is not applied, an additional uncertainty of 0.8% must be assigned. 0.8% of standard uncertainty is estimated by assuming the

Table 1 Physical constants and uncertainties for neutron flux determination [2]

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$c$</td>
<td>0.00112 ± 0.0000036</td>
</tr>
<tr>
<td>$\sigma_0$ (barn)</td>
<td>98.69 ± 0.14</td>
</tr>
<tr>
<td>$\sigma_l$ (barn)</td>
<td>5.57 ± 0.30</td>
</tr>
<tr>
<td>$\sigma_n$ (barn)</td>
<td>1.57 ± 0.30</td>
</tr>
<tr>
<td>$\lambda$ (min)</td>
<td>1.786E-04 ± 0.0005E-04</td>
</tr>
<tr>
<td>$T$ (min)</td>
<td>3879.94 ± 0.43</td>
</tr>
<tr>
<td>$\lambda_{res}$</td>
<td>3879.94 ± 0.43</td>
</tr>
<tr>
<td>$\lambda_{th}$</td>
<td>3879.94 ± 0.43</td>
</tr>
<tr>
<td>$\sigma$ (eV)</td>
<td>0.9554 ± 0.0007 [5]</td>
</tr>
</tbody>
</table>
rectangular distribution of the power ramps correction and the maximum value of the correction is 1.4%. Taking into account of power ramps, the relative uncertainty of the reaction rate becomes 2.0%.

Table 2 shows the uncertainty components and sensitivity coefficients for the reaction rate.

### Table 2: List of uncertainty components for reaction rate (n = 2, net counts = 10,000, t = 1440 min, t = 60 min.)

<table>
<thead>
<tr>
<th>Uncertainty component</th>
<th>Uncertainty type</th>
<th>Relative uncertainty</th>
<th>Sensitivity coefficient</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample mass</td>
<td>B</td>
<td>0.3%</td>
<td>1</td>
</tr>
<tr>
<td>Au concentration</td>
<td>B</td>
<td>0.3%</td>
<td>1</td>
</tr>
<tr>
<td>Net counts rate</td>
<td>A</td>
<td>0.7%</td>
<td>1</td>
</tr>
<tr>
<td>Detection efficiency</td>
<td>B</td>
<td>1.5%</td>
<td>1</td>
</tr>
<tr>
<td>System live time</td>
<td>B</td>
<td>0.2%</td>
<td>1</td>
</tr>
<tr>
<td>Irradiation time</td>
<td>B</td>
<td>0.3%</td>
<td>1</td>
</tr>
<tr>
<td>Power ramp</td>
<td>B</td>
<td>0.8%</td>
<td>1</td>
</tr>
<tr>
<td>Combined uncertainty</td>
<td></td>
<td>2.0%</td>
<td></td>
</tr>
</tbody>
</table>

*The sensitivity coefficient is for the relative uncertainty*

It can be seen from Table 2 that amongst all the uncertainty components the uncertainty of detection efficiency, net count rate and the uncertainty due to power ramps are predominant. Reducing these components can improve the overall uncertainty of reaction rate. To reduce the uncertainty of detection efficiency, a more accurate Au standard in a form of Au-Al alloy wire for the calibration of the HPGe detector is needed. ANSTO is in the process of developing a new method for producing such an accurate standard. For the power ramp, increasing irradiation time from 100 mins to 200 min will reduce the uncertainty components of power ramps from 0.8% to 0.4%. Reduction of the uncertainty of net counts is limited by the fact that during the commissioning stage, there will be large number of samples to be measured in two days. For these reasons it is not practicable to count for long period or have many repeated measurements. However, the minimum 2 repeated counting measurements and 10000 counts per measurement are essential in order to obtain the uncertainty value of 0.7%.

### 3.2 Uncertainty of neutron flux

The uncertainties of conventional neutron flux and epithermal neutron flux represented by the function (7) and (8) may be expressed by (note that for c = 0.00112, Gch =1 and Gres =1. Gch and Gres have negligible uncertainties),

\[
\left( \frac{U_{\phi_0}}{\phi_0} \right)^2 = \left( \frac{1}{R_{cd}} \right)^2 \left( R_{cd}^2 + (1+b)^2 \right) + \left( \frac{U_{R_{cd}}}{R_{cd}} \right)^2 + \left( \frac{1}{U_{I_0}} \right)^2 + b^2 \left( \frac{U_{I_0}}{I_0} \right)^2
\]

and

\[
\left( \frac{U_{\phi_E}}{\phi_E} \right)^2 = \left( \frac{U_{R_{cd}}}{R_{cd}} \right)^2 + \left( \frac{U_{I_0}}{I_0} \right)^2 = 0.028^2
\]

Finally, the uncertainty of thermal neutron flux \( \phi_{th} \) given by the equation (9) is determined by

\[
\left( \frac{U_{\phi_{th}}}{\phi_{th}} \right)^2 = \left( \frac{1}{R_{cd}} \right)^2 \left( R_{cd}^2 + (1+b)^2 \right) + \left( \frac{U_{R_{cd}}}{R_{cd}} \right)^2 + \left( \frac{1}{U_{I_0}} \right)^2 + b^2 \left( \frac{U_{I_0}}{I_0} \right)^2
\]

where, for \( c = 0.00112, a = \frac{1}{G_{th}g\sigma_0} = 0.010081 \) and

\[
\frac{b}{G_{res}I_0} = 0.02732. \quad R_{cd} = \frac{R_s}{R_{s,cd}}
\]

is called cadmium ratio. If the net counts \( C_s \) and \( C_{cd} \) are made approximately equal \( (C_s = C_{cd}) \), we have

\[
\frac{U_{R_s}}{R_s} \approx \frac{U_{R_{cd}}}{R_{s,cd}} \approx \frac{U_{R}}{R} = 2.0%.
\]

The contribution of uncertainty of \( \sigma_0 \) is negligible. The \( w' \) term can be also neglected as \( \frac{w'\sigma_0}{G_{res}I_0} \approx 0 \). The equation (16) and (17) can then be simplified as

\[
\left( \frac{U_{\phi_E}}{\phi_E} \right)^2 = \left( \frac{U_{R_{cd}}}{R_{cd}} \right)^2 + \left( \frac{U_{I_0}}{I_0} \right)^2
\]

and

\[
\left( \frac{U_{\phi_{th}}}{\phi_{th}} \right)^2 = \left( \frac{1}{R_{cd}} \right)^2 \left( R_{cd}^2 + (1+b)^2 \right) + \left( \frac{U_{R_{cd}}}{R_{cd}} \right)^2 + \left( \frac{1}{U_{I_0}} \right)^2 + b^2 \left( \frac{U_{I_0}}{I_0} \right)^2
\]
\[
\left( \frac{U_{\phi_{0.6}}}{\phi_{0.6}} \right)^2 = \frac{1.128^2}{T_0} \left[ \frac{1}{4} \left( \frac{U_T}{T} \right)^2 + \left( \frac{U_{\theta_0}}{\theta_0} \right)^2 \right] + 1.5294 \left( \frac{\phi_T}{\phi_0} \right)^2 \left( \frac{U_{\phi_E}}{\phi_E} \right)^2
\]

or

\[
\frac{U_{\phi_{0.6}}}{\phi_{0.6}} = \sqrt{0.022^2 + 0.005^2 \left( 1 - \frac{1.0273}{R_{cd}} \right)^2 \left( 1 - \frac{0.9409}{R_{cd}} \right)^2}
\]  \hspace{2cm} (20)

In equation (20) and (21), the neutron temperature \( T \) is assumed to be approximately the same as \( T_0 \) and the uncertainty of \( T \) is \( 3^\circ \)K. To make the expression simpler, \( R_{cd} \) is assumed to be greater than 10.

Equation (21) shows that besides the uncertainty components discussed previously in this paper the cadmium ratio \( R_{cd} \) also influences the overall uncertainty of the neutron flux \( \phi_{0.6} \) through the sensitivity coefficient of the reaction rate. When \( R_{cd} > 100 \), the uncertainty of \( \phi_{0.625} \) is regarded as being independent of \( R_{cd} \) and equal to 2.3%.

4. Conclusion

Among the uncertainty components contributing to the overall uncertainty of \( \phi_{0.625} \) determined through activity measurement of a neutron activated Au-Al alloy wire, the uncertainty of 411 keV full-energy peak efficiency, net counts and the uncertainty due to power ramp are the predominant uncertainties. Reduction of these uncertainty components will effectively reduce the overall uncertainty of \( \phi_{0.625} \), and is achievable.

Cadmium ratio \( R_{cd} \) influences the overall uncertainty of the neutron flux \( \phi_{0.625} \) through the sensitivity coefficient of the reaction rate. When \( R_{cd} > 100 \), the uncertainty of \( \phi_{0.625} \) is independent of \( R_{cd} \) and equal to 2.3%.

Reference:


Chapter Eight

Calibration of the Capintec CRC-712M Dose Calibrator for $^{18}$F

L. Mo$^{a,b}$, M.I. Reinhard$^a$, J.B. Davies$^{a,b}$, D. Alexiev$^a$, C. Baldock$^b$

$^a$Australian Nuclear Science and Technology Organisation (ANSTO)
New Illawarra Road, Lucas Heights 2234 NSW Australia

$^b$Institute of Medical Physics, School of Physics, University of Sydney, NSW 2006, Australia

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**Author’s contribution**

L. Mo (candidate)
Designed and carried out experiment, analysed data and wrote manuscript

M.I. Reinhard
Carried out primary standardisation of $^{18}$F and edited manuscript.

J.B. Davies
Checked calculation and edited manuscript.

D. Alexiev
Carried out experiment and edited manuscript.

C. Baldock
Edited manuscript.
Calibration of the Capintec CRC-712M dose calibrator for $^{18}$F

L. Mo$^{a,b,*}$, M.I. Reinhard$^{a}$, J.B. Davies$^{a,b}$, D. Alexieva$^{a}$, C. Baldock$^{b}$

$^{a}$Australian Nuclear Science and Technology Organisation (ANSTO), New Illawarra Road, Lucas Heights, NSW 2234, Australia
$^{b}$Institute of Medical Physics, School of Physics, University of Sydney, NSW 2006, Australia

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Abstract

Primary standardisation was performed on a solution of $^{18}$F using the 4$n$/$\beta$$-\gamma$ coincidence counting efficiency-tracing extrapolation method with $^{60}$Co used as a tracer nuclide. The result was used to calibrate the ANSTO secondary standard ionisation chamber which is used to disseminate Australian activity standards for gamma emitters. Using the secondary activity standard for $^{18}$F, the Capintec CRC-712M dose calibrator at the Australian National Medical Cyclotron (NMC) Positron Emission Tomography (PET) Quality Control (QC) Section was calibrated. The dial setting number recommended by the manufacturer for the measurement of the activity of $^{18}$F is 439. In this work, the dial setting numbers for the activity measurement of the solution of $^{18}$F in Wheaton vials were experimentally determined to be 443$\pm$12, 446$\pm$12, 459$\pm$11, 473$\pm$15 for 0.1, 1, 4.5 and 9 ml solution volumes, respectively. The uncertainties given above are expanded uncertainties ($k = 2$) giving an estimated level of confidence of 95%. The activities determined using the manufacturer recommended setting number 439 are 0.8%, 1.4%, 4.0% and 6.5% higher than the standardised activities, respectively. It is recommended that a single dial setting number of 459 determined for 4.5 ml is used for 0.1–9 ml solution in Wheaton vials in order to simplify the operation procedure. With this setting the expended uncertainty ($k = 2$) in the activity readout from the Capintec dose calibrator would be less than 6.2%.

Keywords: $^{18}$F; Solution; Activity measurement; Dose calibrator; Ionisation chamber; Dial setting number

1. Introduction

Cyclotron produced $^{18}$F in the form of 2-[18F]-fluoro-2-deoxyglucose [($^{18}$F)FDG)] is mainly used in positron emission tomography (PET) imaging for assessment of abnormal glucose metabolism to assist in the diagnosis of cancer. Accurate administration of radiopharmaceuticals for diagnosis in nuclear medicine requires the instrument for activity measurement, the dose calibrator, to be calibrated and traceable to national standards.

The Australian National Medical Cyclotron (NMC) routinely produces ($^{18}$F)FDG. The ionisation chamber used by the NMC PET Quality Control (QC) Section is the commercial re-entrant dose calibrator, model CRC-712M, manufactured by Capintec Inc (Pittsburgh, PA, USA). The activity values indicated by the instrument depend on the induced ionisation current and a conversion factor in the form of a dial setting number that converts pA to MBq. The instrument manufacturer has determined the dial setting numbers from calibration curves obtained by measuring the response of the chamber to $^{57}$Co and $^{60}$Co (Capintec, Inc., 1990). The dial setting number that is recommended by Capintec for $^{18}$F is 439. This is only valid for the geometry of 5 ml of solution in a 5 ml ampoule.

Investigation of dial settings of Capintec dose calibrators has been performed on various geometries on $^{90}$Y (Coursey et al., 1993) and $^{117m}$Sn, $^{62}$Cu, $^{188}$Re, $^{125}$I, $^{99m}$Tc, $^{133}$Xe, $^{153}$Gd, $^{153}$Sm and $^{18}$F (Zimmerman et al., 1998, 1999, 2001; Zimmerman and Cessna, 1999, 2000). It was found that most of the dial settings recommended by the manufacturer result in an overestimation of the activity by up to 50%. In the case of $^{18}$F, up to 6% of overestimation of activity was found depending on the sample geometries.

$^{18}$F decays by positron emission with an intensity of 96.86% and electron capture with an intensity of 3.14% to...
the ground state of stable nuclide $^{18}$O. The half-life of $^{18}$F is 1.8288 h (Be et al., 2004). The positron has a maximum energy of 633.5 keV and an average energy of 249.3 keV. The 511 keV annihilation radiation following the emission of a positron has an intensity of 193.72% (Be et al., 2004). Liquid scintillation counting with the CIEMAT/NIST and TDCR methods and $4\pi \beta-\gamma$ coincidence counting technique have been used to standardise this radionuclide (Woods and Baker, 2004; Ratel et al., 2005).

In this work, the primary standardisation of $^{18}$F was performed using the $4\pi \beta-\gamma$ coincidence counting efficiency-tracing extrapolation method with $^{60}$Co as the tracer. The standardised solution was then used to calibrate the ANSTO secondary standard ionisation chamber. The ANSTO Secondary Standard ionisation chamber is a re-entrant, high-pressure TPA ionisation chamber (Sharpe and Wade, 1951). It is used as a working chamber for the routine dissemination of the activity standards for photon emitting radionuclides. The CRC-712M dose calibrator located at the NMC PET QC section was calibrated for the geometry of 0.1, 1, 4.5 and 9 ml solutions in 10 ml Wheaton vials and the standard activity of the solution was determined using the ANSTO secondary standard ionisation chamber.

2. Experimental

A solution of ($^{18}$F)FDG in an isotonic solution of sodium chloride was obtained from the NMC. The solution was diluted by the addition of 0.1 M HCl to produce the F-18 master solution. The master solution had an activity concentration of approximately 980 kBq/g in 12 ml. Nominally 0.8 g of the $^{18}$F solution was taken from the master solution and mixed with the nominally 0.2 g of the $^{60}$Co solution in a vial. From the mixture, a set of six $4\pi$ counting sources was prepared on Pd/Au plated VYNS source mounts for the primary standardisation to determine the activity concentration of the master solution. The exact mass dispensed was determined by weighing the syringe pycnometer before and after dispensing. Another set of three samples was prepared with nominally 3.6 g taken from the master solution and dispensed into the 5 ml BIPM standard glass ampoules for the calibration of the ANSTO secondary standard ionisation chamber. The mass dispensed from the master solution into the ampoules was determined by weighing the ampoules together with the cap before and after dispensing. The ampoules were then flame-sealed.

The primary standardisation was performed using a conventional $4\pi \beta/(PC)–\gamma$ coincidence counting system. A $4\pi$ gas-flow proportional counter was used for the $\beta$ detection and a NaI(Tl) scintillator used for the $\gamma$ detection. The $4\pi \beta-\gamma$ coincidence counting efficiency-tracing extrapolation method (Baerg, 1973) was applied and $^{60}$Co was used as the tracer. Al foils were used to attenuate the $\beta^-$ from the $^{18}$F and $\beta^+$ from the $^{60}$Co in order to vary the $\beta$ detection efficiency. The activity of $^{18}$F was determined by the subtraction of the tracer activity from the extrapolated absolute total $\beta$ count rate of the mixture of $^{18}$F and $^{60}$Co radionuclides. The activity concentration of the master solution was then obtained and used to calibrate the ANSTO Secondary Standard ionisation chamber. Radium check sources were used in all secondary standard ionisation chamber measurements to correct for the instability of the ionisation chamber and the inaccuracy of the electrometer. The details of the primary standardisation procedures are described by Reinhard et al. (2002).

A second batch of ($^{18}$F)FDG stock solution was obtained from the NMC for the calibration of CRC-712M dose calibrator at the NMC at a later time. The stock solution was diluted with 0.1 M HCl to produce a 15 ml master solution with an activity of approximately 20 GBq. The master solution was then dispensed into a series of 0.1, 1, 4.5 and 9 ml solutions in Wheaton vials.

The samples were measured in the dose calibrator in the same position with the readout of the Capintec dose calibrator displayed in units of activity. Measurements were made as a function of the dial setting from 420, in increments of 10, to 500. A radium check source was used for stability measurements.

The samples were then dispensed into 5 ml BIPM standard glass ampoules with nominally 3.6 ml of ($^{18}$F)FDG solution and the activity determined using the secondary standard ionisation chamber.

Detection of photon emitting impurities was carried out using a HPGe crystal detector (Model no.: GMX-30190-P). The crystal dimension was 54.8 mm in diameter and 76.8 mm in length. The source was positioned at 90 mm away from the detector end cap (93 mm from crystal head). The signals from the detector were processed by the ORTEC 92X Spectrum MASTER™. The software used was ORTEC MAESTRO™ for Windows.

The linearity of both secondary standard ionisation chamber and CRC-712M dose calibrator was checked by measuring the response to a $^{99m}$Tc source over eight half-lives.

The “calibration curve method” (Zimmerman and Cessna, 2000) was employed for the experimental determination of the dial setting number. It is described below.

For each sample, the dose calibrator activity readout $A_{\text{cap}}$ was divided by the standard activity $A_{\text{std}}$ estimated by the secondary standard ionisation chamber, giving the ratio $R = A_{\text{cap}}/A_{\text{std}}$. The values of $R$ vs dial setting number were then fitted to the equation,

$$R^{-1} = a \times \text{(Dial setting)} + b. \quad (1)$$

Solving the fitting equation for $R = 1$ enables the determination of the experimental dial setting number for each sample.

The uncertainties were evaluated in accordance with the ISO GUM (International Organisation for Standardisation, 1993) and are quoted here as standard uncertainties with a coverage factor of $k = 1$ providing a confidence level of 68% unless otherwise specified.
3. Results and discussion

It is noted that the impurity of $^{48}$V was found in the ($^{18}$F)FDG produced at the PET Center of the National Institutes of Health in the USA (Zimmerman et al., 2001). However, neither this work, nor the NMC during their 12 years of production of ($^{18}$F)FDG detected any $\gamma$-emitting impurities above the detection limit of the $\gamma$-ray spectroscopy system.

The activity concentration of the ($^{18}$F)FDG primary solution determined using the $4\pi \beta - \gamma$ coincidence counting efficiency-tracing extrapolation method was found to be $1144.8 \pm 1.4$ % at the reference time. The uncertainty components for the activity concentration of the primary source of ($^{18}$F)FDG are given in Table 1. It can be seen that the dominant contributors were the uncertainty in the efficiency extrapolation (0.90%) and the standard deviation of the samples (0.92%). The accuracy of the primary standardisation was confirmed through an international comparison of the activity of $^{18}$F coordinated by the National Physical Laboratory in the UK (Woods and Baker, 2004). ANSTO’s result differed from the key comparison reference value by $-0.60\%$ (Ratel et al., 2005).

A calibration factor of 29.774 pA/MBq $\pm 1.4\%$ was determined for the ANSTO Secondary Standard ionisation chamber for the geometry of 3.6 ml solution in a 5 ml ampoule. The uncertainty components for the calibration factor are given in Table 2. The dominant uncertainty contributor was the activity concentration of the primary solution (1.4%). The other contributions listed in the Table 2 were insignificant in comparison with the large uncertainty in the activity concentration of the primary solution.

The standard activity concentration of the second batch of ($^{18}$F)FDG determined by the secondary standard ionisation chamber was $1334.82 \pm 1.5$ % at the reference time. The ratio of the standard activity $A_{std}$ to the activity readout from the Capintec CRC-712 M dose calibrator $A_{cap}$, $R^{-1} = A_{std}/A_{cap}$, was plotted against the dial setting number and fitted to Eq. (1) using Microsoft Excel™. The corrected dial setting for each volume was determined by taking an average of two samples. See Table 3 for the determined dial setting number and its uncertainty for each sample volume.

The values for $R = A_{cap}/A_{std}$ have a standard uncertainty of 1.6%. The uncertainty components contributed to the combined uncertainty in $R$ are listed in Table 4. The calibration factor for the secondary standard ionisation chamber contributed 1.4% of the combined uncertainty in $R$-value. The calibrated current of radium check source for the secondary standard ionisation chamber contributed 0.30% and the activity of the radium check source for the dose calibrator contributed 0.40%. The non-linearity for the Secondary Standard ionisation chamber and dose calibrator contributed 0.35% and 0.50% respectively. The variations in wall thickness of the Wheaton vial contributed $0.30\%$. The other uncertainty components have minimal influence. The standard uncertainty in the dial setting was determined by solving the fitting Eq. (1) for the dial setting values for $R = 1 \pm U_R$, using the approach of Zimmerman et al. (2001).

### Table 1

Uncertainty components for the activity concentration of the primary source of ($^{18}$F)FDG standardised using the $4\pi \beta - \gamma$ coincidence counting efficiency-tracing extrapolation method

<table>
<thead>
<tr>
<th>Source of uncertainty</th>
<th>Standard uncertainty components (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Type A</td>
</tr>
<tr>
<td>Counting statistics</td>
<td>0.17</td>
</tr>
<tr>
<td>Dead time</td>
<td></td>
</tr>
<tr>
<td>Activity concentration of $^{60}$Co tracer</td>
<td>0.38</td>
</tr>
<tr>
<td>Mass of 4$\pi$ counting source</td>
<td></td>
</tr>
<tr>
<td>Mass of $^{18}$F and $^{60}$Co mixture</td>
<td>0.03</td>
</tr>
<tr>
<td>Mass of $^{18}$F master solution</td>
<td>0.04</td>
</tr>
<tr>
<td>Half life of $^{18}$F</td>
<td>0.04</td>
</tr>
<tr>
<td>$^{18}$F $\beta^+$ emission probability</td>
<td></td>
</tr>
<tr>
<td>Intercept from fitted curve</td>
<td>0.90</td>
</tr>
<tr>
<td>Standard deviation of samples</td>
<td>0.92</td>
</tr>
<tr>
<td>Quadratic sum</td>
<td>1.4</td>
</tr>
<tr>
<td>Combined standard uncertainty</td>
<td></td>
</tr>
</tbody>
</table>

### Table 2

Uncertainty components for the calibration factor for ($^{18}$F)FDG for the ANSTO secondary standard ionisation chamber

<table>
<thead>
<tr>
<th>Source of uncertainty</th>
<th>Standard uncertainty components (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Type A</td>
</tr>
<tr>
<td>Activity concentration of primary solution</td>
<td></td>
</tr>
<tr>
<td>ESDM$^a$ of sample current</td>
<td>0.05</td>
</tr>
<tr>
<td>ESDM of background current</td>
<td>0.02</td>
</tr>
<tr>
<td>ESDM of Ra check source current at measurement time</td>
<td>0.03</td>
</tr>
<tr>
<td>Ra check source current at calibration time $t_c$</td>
<td>0.30</td>
</tr>
<tr>
<td>Non-linearity of the chamber</td>
<td>0.35</td>
</tr>
<tr>
<td>Half life of $^{18}$F</td>
<td>0.02</td>
</tr>
<tr>
<td>Quadratic sum</td>
<td>0.36</td>
</tr>
<tr>
<td>Combined standard uncertainty</td>
<td></td>
</tr>
</tbody>
</table>

$^a$Experimental standard deviation of the mean.

### Table 3

Determined dial setting number and its uncertainty for different geometries

<table>
<thead>
<tr>
<th>Volume (ml)</th>
<th>Dial setting</th>
<th>Uncertainty$^b$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.1</td>
<td>443</td>
<td>12</td>
</tr>
<tr>
<td>1</td>
<td>446</td>
<td>12</td>
</tr>
<tr>
<td>4.5</td>
<td>459</td>
<td>11</td>
</tr>
<tr>
<td>9</td>
<td>473</td>
<td>15</td>
</tr>
</tbody>
</table>

$^b$The uncertainty is an expanded uncertainty with coverage factor $k = 2$ giving 95% confidence level.
Although the international comparison of the activity of $^{18}$F confirmed the accuracy of ANSTO’s primary standardisation of the nuclide, $^{60}$Co might not be an ideal tracer to be used for the absolute measurement of activity of $^{18}$F. The uncertainty in the activity concentration determined by the tracing method using $^{60}$Co as a tracer was as large as 1.4%, mainly due to the large uncertainty in the efficiency extrapolation and standard deviation of different samples. With 1.4% standard uncertainty being propagated to the calibration factor and then the standard activity value determined using the secondary standard ionisation chamber, a large uncertainty was consequently obtained for the dial setting. This is the reason why the expanded uncertainties in the dial setting in this work were larger by a factor of two compared with the work of Zimmerman et al. (2001).

The dial setting recommended by the manufacturer, and currently used by the NMC, is 439 which is lower than the dial setting determined in this work resulting in overestimation of sample activities by 0.8%–6.5% for the sample volume of 0.1–9 ml. Use of a larger volume results in a greater discrepancy.

The experimental results showed that if the dial setting of 459 for 4.5 ml was used for the 0.1, 1, and 9 ml solutions, the $R$-value would be 1.030, 1.025, 1.025. This would give rise to an uncertainty of ≤3% for all volumes of solution investigated. However, even with consideration of other uncertainty sources such as the linearity of the dose calibrator (0.50%), stability correction for the dose calibrator using the radium check source (0.40%) and the variation of wall thickness of the Wheaton vials (0.30%), the combined standard uncertainty would be 3.1% (or the expanded uncertainty of 6.2% with $k = 2$ for an estimated level of confidence of 95%) for the activity readout from the dose calibrator. In the nuclear medicine environment, as the “rule of thumb”, the acceptable tolerance of the error for activity determination is 10%. Therefore, if a single dial setting is required for any volume between 0.1 and 9 ml determined in Wheaton vials, a dial setting of 459 would be recommended.

When the dial setting was plotted against solution volume, it was found that the dial setting increased linearly with sample volume (Fig. 1).

For photon-emitting nuclides one would expect that the dial setting [or calibration factor (pA/MBq)] would increase as the sample volume decreases. The opposite trend, observed in Fig. 1, indicates that the dial settings were affected by the variations in geometry within the chamber well. The relatively high-energy (511 keV) annihilation photons may not significantly affect the dial setting with changing solution volume.

### Table 4

<table>
<thead>
<tr>
<th>Source of uncertainty</th>
<th>Standard uncertainty components (Type A)</th>
<th>(%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Calibration factor for the secondary standard chamber</td>
<td></td>
<td>1.4</td>
</tr>
<tr>
<td>ESDM of sample current in secondary standard chamber</td>
<td></td>
<td>0.04</td>
</tr>
<tr>
<td>ESDM of background current</td>
<td></td>
<td>0.02</td>
</tr>
<tr>
<td>ESDM of Ra check source current at measurement time $t_m$</td>
<td></td>
<td>0.03</td>
</tr>
<tr>
<td>Ra check source (for secondary standard chamber) current at Ra calibration time $t_c$</td>
<td></td>
<td>0.30</td>
</tr>
<tr>
<td>Non-linearity of the secondary standard chamber</td>
<td></td>
<td>0.35</td>
</tr>
<tr>
<td>Non-linearity of the dose calibrator</td>
<td></td>
<td>0.50</td>
</tr>
<tr>
<td>Activity of Ra check source (for dose calibrator)</td>
<td></td>
<td>0.40</td>
</tr>
<tr>
<td>ESDM of activity readout from dose calibrator</td>
<td></td>
<td>0.07</td>
</tr>
<tr>
<td>Sample mass</td>
<td></td>
<td>0.05</td>
</tr>
<tr>
<td>Variation in wall thickness of Wheaton vial</td>
<td></td>
<td>0.30</td>
</tr>
<tr>
<td>Half life of $^{18}$F</td>
<td></td>
<td>0.08</td>
</tr>
<tr>
<td>Quadratic sum</td>
<td></td>
<td>0.62</td>
</tr>
<tr>
<td>Combined standard uncertainty</td>
<td></td>
<td>1.6</td>
</tr>
</tbody>
</table>

![Fig. 1. Plot of dial setting number as function of solution volume. The solid line is a linear fit.](image)

Fig. 1. Plot of dial setting number as function of solution volume. The solid line is a linear fit.

4. Conclusion

Primary standardisation was carried out on a solution of $^{18}$FFDG using the $4\pi\beta-\gamma$ coincidence counting efficiency-tracing extrapolation method. The result agreed with the key comparison reference value within 0.60% as verified in an international comparison. The result was used to calibrate the ANSTO secondary standard ionisation chamber.
Using the secondary activity standard for the (18F)FDG, the Capintec CRC-712M dose calibrator located at the NMC PET QC section was calibrated. New dial setting numbers, 443 ± 12, 446 ± 12, 459 ± 11, 473 ± 15, for the Capintec dose calibrator have been experimentally determined for (18F)FDG for the 0.1, 1, 4.5 and 9 ml solutions in a Wheaton vial, respectively. To simplify the operation procedure, a dial setting of 459 for 4.5 ml is recommended for any volume between 0.1 and 9 ml solution in Wheaton vials. The uncertainty in the activity readout from the Capintec dose calibrator by using a single dial setting is well below 10%, an acceptable error tolerance of activity measurement in the nuclear medicine environment.

Acknowledgments

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References


Chapter Nine

Development of an Australian secondary standard for the reference air kerma rate measurement of $^{125}$I seeds

J.B. Davies, L. Mo, D. Alexiev

Australian Nuclear Science and Technology Organisation (ANSTO), New Illawarra Road, Lucas Heights 2234 NSW Australia

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Author’s contribution

J.B. Davies
Designed and carried out experiment, analysed data and wrote manuscript.

L. Mo (candidate)
Designed experiment and edited manuscript.

D. Alexiev
Contributed to manuscript editing.
TECHNICAL NOTE

Development of an Australian secondary standard for the reference air kerma rate measurement of $^{125}$I seeds

J. B. Davies, L. Mo and D. Alexiev

Safety & Radiation Science, Australian Nuclear Science & Technology Organisation, Menai, Australia

Abstract

Calibration of a High-Dose-Rate 1000 Plus ionisation chamber and associated electrometer for the determination of reference air kerma rate for $^{125}$I brachytherapy seeds has been developed. Traceability was established from ionisation chamber measurements of an Amersham model 6711 $^{125}$I seed for which a primary standard measurement of reference air kerma rate has been made by the National Institute of Standards and Technology. The status of an Australian secondary standard is to be realised by establishing legal verifying authority from the National Measurement Institute. The calibrated chamber thus provides a measurement from which similar chambers in hospitals may be calibrated. For quality assurance this result was compared with the calibration certificate supplied by the Accredited Dosimetry Calibration Laboratory of the University of Wisconsin, USA.

Key words brachytherapy, $^{125}$I, calibration, dosimetry, ionisation chamber

Introduction

For specification of the photon sources used in brachytherapy, IAEA-TECDOC-1274 recommends the ICRU quantity “reference air kerma rate”, $K_R$, defined as the air kerma rate at a reference distance of one metre, after correction for air attenuation and scattering. It is recommended this quantity be expressed in $\mu$Gy.h$^{-1}$ for LDR brachytherapy applications. Effective brachytherapy treatment relies on measurements of reference air kerma rate being traceable to standards of the highest metrological quality. In the case of $^{125}$I, the only established primary standard for low energy photon sources is held by the National Institute of Standards and Technology (NIST), USA, and until now, there has not been a local system for dissemination of the NIST standard.

This work establishes a well type ionisation chamber based system for $^{125}$I reference air kerma rate measurement with direct traceability to the NIST primary standard. This was achieved by sending a single $^{125}$I seed to NIST for reference air kerma rate measurement and then using that seed to determine a traceable calibration coefficient for the ANSTO ionisation chamber.

Material and methods

The brachytherapy seed measurement system at ANSTO consists of a well type Standard Imaging High-Dose-Rate (HDR) DR 1000 Plus ionisation chamber with source holders designed for insertion of brachytherapy seeds. The chamber is connected to a calibrated Keithley 3540 electrometer from which a bias of +300V is applied. The charge collected is negative. $^{241}$Am and $^{137}$Cs check sources mounted within custom-made source holders are used for ionisation chamber stability checks (Figure 1).

Stability and linearity

Ionisation chamber stability checks were done using $^{241}$Am and $^{137}$Cs sources mounted within custom-made source holders. The use of a $^{137}$Cs source for stability checks was done for consideration of future work with $^{192}$Ir. During this calibration period, the average check source measurement values, corrected for decay, were:

- $I_{^{241}Am} = -3.15 \pm 0.05$ pA
- $I_{^{137}Cs} = -48.39 \pm 0.66$ pA

The uncertainties are quoted at a level of confidence of 95% using $k = 2$ (uncertainties are discussed later). IAEA-TECDOC-1274 recommends criteria within which it can be assumed the calibration has not changed if the stability checks are within¹.
Development of an Australian secondary standard

Figure 1. Clockwise from top left: the HDR1000+ ionisation chamber, Keithley 35040 and source holders.

Figure 2. Linearity of the HDR 1000+ ionisation chamber.

± 1.5% for low energy photon sources (e.g. $^{241}$Am); or ± 1% for high energy photon sources (e.g. $^{137}$Cs).

The maximum deviation of the stability check source measurements from the mean was 1.3% for $^{241}$Am and 0.96% for $^{137}$Cs.

The linearity of the dosimetry system was tested by comparison of the measured response to that expected allowing for radioactive decay. A 5ml solution of approximately 5GBq of $^{99m}$Tc was used for this purpose. Figure 2 shows the variance of linearity relative to the initial measurement of 204 pA. A positive current was measured in this case. The combined standard uncertainty was calculated taking into account air density and radioactive decay corrections for each reading and is indicated as error bars in Figure 2.

Given that the current measured for the $^{125}$I seeds was approximately –25 pA, an uncertainty of 0.5% with a rectangular distribution was estimated in the linearity of this ionisation chamber.

Calibration methodology

The calibration coefficient developed was determined based on the NIST measurement of reference air kerma rate of an $^{125}$I seed (Amersham model 6711) and ANSTO measurements of current from the same seed using the measurement system shown in Figure 1.

Seed measurements

Prior to any measurement, the ion chamber was connected to the electrometer, bias applied and the instrument given time to stabilise. It was demonstrated that stability of the system required a minimum of fifteen minutes.

Four Amersham model 6711 $^{125}$I seeds were obtained from Royal Perth Hospital. A seed was inserted into a Teflon tube housed inside the LDR Model 70016 single seed source holder. The seed was positioned vertically in the holder in a stable geometry.

Charge and time measurements were taken from the Keithley electrometer. Multiple measurements and source insertions of each seed were performed.

Every individual reading was corrected to reference conditions. Since this type of chamber is open to the atmosphere corrections must be made for changes in air density. The reference conditions used were 20°C and 101.325kPa. The devices used to measure temperature (mercury thermometer) and pressure (Aneroid barometer) have measurement traceability to the relevant Australian standards. No correction was made for relative humidity. Corrections were made for the decay of $^{125}$I using a half-life of 59.408 days. The effect of ion recombination was determined to be less than 0.1%. A correction for ion recombination was not applied but an uncertainty component of 0.1% was calculated and included in the overall measurement uncertainty.

Gamma spectrometry

The photon emission spectrum of another $^{125}$I seed from the same batch was analysed to assess the purity of the source. The activity of the seed was too high to allow a spectrum measurement at the time of calibration, so this measurement was performed several months after the ionisation chamber measurement. Thus the possibility of short-lived impurities remains indeterminate. A HP germanium detector with a 0.125mm thick Be window was used to search for low energy photons (<60keV). A second HP germanium detector with a 0.5mm thick Be window and its end cap in place was used to search for higher energy photons.

Results and discussion

No high energy photons were detected in the gamma-ray spectrum. No long-lived gamma-emitting impurities were found in this $^{125}$I seed. Low energy x-rays (27.4 - 31.7keV) and a 35.5keV $\gamma$-ray attributed to $^{125}$I were detected. Ag fluorescence x-rays (22.1keV and 24.9keV) from the Model 6711 seed’s silver wire construction were also detected, and shown in Figure 3.

The results of ionisation chamber measurements, corrected as described above, are displayed in Figure 4. The error bars shown are the Type A uncertainty, indicating the precision only in the ionisation chamber measurement.
Figure 3. Measured photon spectrum of an 125I Model 6711 seed.

Figure 4. Corrected current (pA) for measurement of four 125I seeds.

Table 1. Calibration uncertainty budget.

<table>
<thead>
<tr>
<th>Quantity</th>
<th>Relative standard uncertainty, u_i (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Type A</td>
</tr>
<tr>
<td>Reproducibility in current:</td>
<td>0.30</td>
</tr>
<tr>
<td>Electrometer accuracy in charge measurement:</td>
<td>0.20</td>
</tr>
<tr>
<td>Exposure period:</td>
<td>0.06</td>
</tr>
<tr>
<td>Temperature:</td>
<td>0.10</td>
</tr>
<tr>
<td>Pressure:</td>
<td>0.01</td>
</tr>
<tr>
<td>125I half-life:</td>
<td>0.0005</td>
</tr>
<tr>
<td>Linearity:</td>
<td>0.29</td>
</tr>
<tr>
<td>Long term stability:</td>
<td>0.46</td>
</tr>
<tr>
<td>Ion recombination:</td>
<td>0.10</td>
</tr>
<tr>
<td>NIST reference air kerma rate:</td>
<td>0.88</td>
</tr>
<tr>
<td>Quadratic sum</td>
<td>0.30</td>
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</table>

Relative combined standard uncertainty in $N_i$: 1.106
Effective degrees of freedom, $v_{eff}$: 473
Relative expanded uncertainty in calibration coefficient, $N_i$: 2.21

Calibration coefficient

The reference air kerma rate calibration coefficient for the model 6711 seed, $N_i$, for this chamber was calculated from the quotient of the NIST reference air kerma rate, $K_R$, (in units of $\mu$Gy.h$^{-1}$) and the average corrected ionisation chamber current (pA).

The NIST reference air kerma rate, $K_R$, corrected for decay, was:

$$K_R = 5.76 \pm 0.10 \ \mu\text{Gy.h}^{-1}$$

Subsequently, $N_i$, becomes:

$$N_i = -0.2334 \pm 0.0052 \ \mu\text{Gy.h}^{-1}.\text{pA}^{-1}$$

The negative sign indicates a negative charge measured by the electrometer.

Comparison to previous calibration

The Accredited Dosimetry Calibration Laboratory (ADCL) of the University of Wisconsin, USA, provided a calibration certificate with traceability to the primary standard at NIST for the ionisation chamber for 125I and 192Ir seeds in June 2003. For quality assurance the result of this work was compared with the 2003 ADCL calibration.

The calibration coefficient reported by the ADCL for seed model 6711 was referenced to 22°C and 760 Torr:

$$N_i = 0.2334 \pm 0.0058 \ \mu\text{Gy.h}^{-1}.\text{pA}^{-1}$$

The positive sign indicates a positive charge measured by the ADCL electrometer.

To compare this with the result of this work, the above $N_i$ from ADCL must be converted to a reference temperature of 20°C. The ADCL calibration coefficient at 20°C becomes:

$$N_i = 0.2318 \pm 0.0058 \ \mu\text{Gy.h}^{-1}.\text{pA}^{-1}$$

Thus the result of this work differs from the ADCL calibration by 0.69%. This is well within the uncertainty of the new calibration coefficient of 2.21%.

Uncertainties

All uncertainties stated are the expanded uncertainties to a level of confidence of approximately 95%, unless specified otherwise. The expanded uncertainty was obtained by multiplying the combined standard uncertainty by a coverage factor of two. In determining the calibration coefficient, the uncertainty of each influence quantity is given as a standard uncertainty relative to the calibration coefficient. The calculation follows the ISO Guide for the expression of uncertainty in measurements.

A breakdown of the calculation of uncertainties for this calibration is given in Table 1.

Conclusion

Calibration of an ionisation chamber for the reference air kerma rate measurement of 125I seeds was established, with traceability to the NIST primary standard measurement. This result was found to differ from the ADCL calibration in June 2003 by 0.69%. The calibrated system developed in this work may be used to establish traceable reference air kerma rate measurements.
Acknowledgments

The authors would like to acknowledge Dr Leonard Wee and colleagues of the Royal Perth Hospital for supplying the $^{125}$I seeds used in this work.

References

Chapter Ten

Comparison of LaBr$_3$·Ce, LaCl$_3$·Ce, CZT and NaI(Tl)
for Resolution of Nuclear Material Spectra

D. Alexiev$^a$, L. Mo$^{a,b}$ and M.L. Smith$^a$

$^a$Australian Nuclear Science and Technology Organisation (ANSTO), PMB1, Menai, NSW, Australia

$^b$Institute of Medical Physics, School of Physics, University of Sydney, NSW 2006, Australia

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Author’s contribution

D. Alexiev
Designed experiment, analysed data and wrote manuscript.

L. Mo (candidate)
Conducted experiment, analysed data and edited manuscript.

M.L. Smith
Contributed to data analysis and manuscript.
Comparison of LaBr$_3$:Ce, LaCl$_3$:Ce, CZT and NaI(Tl) for Resolution of Nuclear Material Spectra

D. Alexiev$^a$, L. Mo$^{ab}$ and M. L. Smith$^a$

$^a$Australian Nuclear Science and Technology Organisation, New Illawarra Road, Lucas Heights, NSW 2234, Australia, $^b$Institute of Medical Physics, School of Physics, University of Sydney, NSW 2006, Australia

Abstract

Energy resolution and efficiency comparison were made of new scintillators, Lanthanum Bromide (LaBr$_3$:Ce), Lanthanum Chloride (LaCl$_3$:Ce), with Cadmium Zinc Telluride (CdZnTe or CZT) and Sodium Iodide (NaI(Tl)). The experiments conducted have shown that LaBr$_3$:Ce, and LaCl$_3$:Ce scintillator crystal provided by Saint-Gobain have adequate resolution and efficiency for measurement of spectral response in the region of 150 keV to 450 keV. An overall advantage in energy resolution and efficiency of LaBr$_3$:Ce and LaCl$_3$:Ce over NaI(Tl) and CZT has also been demonstrated.

Keywords: LaCl$_3$:Ce, LaBr$_3$:Ce, CZT, NaI(Tl), gamma spectral comparison, resolution, efficiency.

1. Introduction

New scintillators, activated with cerium (Ce$^{3+}$) atoms as the primary scintillation source such as LaBr$_3$:Ce and LaCl$_3$:Ce have gained interest [1] due to good resolution, high efficiency and fast response [2] with high light output per keV [3]. LaBr$_3$:Ce and LaCl$_3$:Ce are doped with Ce to increase the photons per keV light output and to reduce the their decay time [4]. These two properties combine to classify this material as a possible radiation detector which could replace CZT and NaI(Tl) for a variety of applications requiring room - temperature - operation detectors.

In this work comparisons are made of key parameters such as energy resolution and detection efficiency of LaBr$_3$:Ce and LaCl$_3$:Ce with NaI(Tl) and CdTeZn detectors. The motivation of this study is to evaluate LaBr$_3$:Ce and LaCl$_3$:Ce scintillators for application in Ionising Radiation Physics, such as coincidence counting of beta – gamma emitters [5], activity measurement for neutron flux determination [6] and Safeguards applications [7], for example, the resolving of the key gamma - emission energies of: $^{235}$U, highly enriched uranium (HEU), and $^{239}$Pu. The application of these detectors can be: border monitoring and shipment - container surveying. High spectral resolution and high efficiency are desirable for the identification of shipments containing high quantities of naturally occurring radioactive materials (NORM); for example shipments of some sands, ceramics glaze, fertiliser sand other shipments containing radioactive materials such as
nuclear, industrial, medical and illegally deemed radioactive sources. These scintillators and the CZT detector are tested by comparing the resolution of different detectors, in the energy range of 0.2 to 1.2 MeV, using NORM isotopes and observing separation and their identification.

2. Experimental

A LaBr$_3$:Ce and a LaCl$_3$(Ce) scintillator, complete with a photomultiplier tube and a three stage emitter follower - preamplifier was obtained from Saint-Gobain [8]. Both scintillators have a diameter 25.4 mm and length of 25.4 mm, and are coupled to a Photonis XP2060B ten - stage photomultiplier tube (gain of 6.5x10$^7$ [9] ). This type of photomultiplier may have a gain that is too large for the number of high energy gamma output photons, and consequently may saturate. A more suitable photomultiplier [10] could be the Photonis XP3212B; this photomultiplier tube has eight stages and a gain of 2.5x10$^5$. We note in the published data by Saint-Gobain that the light output of LaBr$_3$:Ce and LaCl$_3$:Ce is 49 photons/keV, compared with 38 photons/keV for NaI(Tl). Moreover, the decay time constant of the LaBr$_3$:Ce and LaCl$_3$(Ce) light pulse has an order of 28 ns, again compared with 250 ns for the NaI(Tl) light pulse. Thus there are special requirements concerning the linearity of the current output of the photomultiplier for LaBr$_3$:Ce and LaCl$_3$:Ce. Its peak current is an order of magnitude larger than that of NaI(Tl). The radionuclides used in the experiment were $^{241}$Am, $^{133}$Ba, $^{152}$Eu, $^{137}$Cs, $^{60}$Co, $^{219}$Pu, HEU ($^{235}$U, 97% enriched), $^{266}$Ra and $^{232}$Th. These isotopes were used in order to give a reasonable overall energy range and for a number of energies within both the region of interest (approximately 100 to 450 keV) and for overall performance evaluation.

The CZT (CdTeZn) detector, type CZT/500, was supplied by RITEC. The NaI(Tl) scintillator assembly, model 30B30/1.5-HV-E3-X, was supplied by AMP-TEK. All radionuclides used were ‘point sources’, supplied by ORTEC model RSS – 8. The actinide used in this report was available in the laboratory. All radioisotopes were placed on a lucid assembly, 10cm central in front of the scintillator assembly or the CZT detector surface located in a lead castle.

3. Results

3.1 Intrinsic Spectral Analysis

Both LaBr$_3$:Ce and LaCl$_3$:Ce contains natural Lanthanum (La), consisting of approximately 99.9% stable $^{139}$La, and approximately 0.09% unstable impurity $^{138}$La, which decays with a half-life of 1.05x10$^{11}$ years. It undergoes 66.4% electron capture to $^{138}$Ba (which is a stable isotope) and
33.6% beta decay to $^{138}\text{Ce}$ (stable cerium and the dopant used for LaBr$_3$ and LaCl$_3$). This causes the detector to be slightly self active which can be seen as spurious spectral lines when collecting a spectrum.

Both LaBr$_3$:Ce and LaCl$_3$:Ce were measured with a HPGe detector in a low background lead castle, each over a period of 4 days, with the background subtracted. The intrinsically produced photons are observed in Figure 1.

![LaCl$_3$:Ce intrinsic spectrum with background subtracted count in a lead castle (taken with a HPGe detector).](image)

Figure 1. LaCl$_3$:Ce intrinsic spectrum with background subtracted count in a lead castle (taken with a HPGe detector).

The spectrum in Figure 1 shows the three intrinsic characteristic spectral lines of $^{138}\text{La}$: 788.7 keV and 1435.8 keV gamma lines as well as low energy X-ray fluorescence peaks from the 32.194 keV K$\alpha$1 and 31.817 keV K$\alpha$2.

A 'self' activation spectrum was also taken of the LaCl$_3$:Ce detector over four days with background subtracted in a low background lead castle to illustrate the internal (self) detection of the intrinsic decay spectrum of $^{138}\text{La}$, shown in Figure 2.
Figure 2. LaCl$_3$:Ce intrinsic internal decay spectrum, 4 day count with background subtraction.

Observed in the internal background spectrum is the beta decay continuum with an $E_{\text{max}}$ at approximately 260 keV. This consisting of 33.6% beta decay with $E_{\text{max}}$ of 255.26 keV and 66.6% electron capture with $E$ of 302.18 keV. An emission line at about 1460 keV corresponds to the overlapped 1460 keV line of $^{40}$K originating from the glass of the photomultiplier tube and that of 1435 keV of $^{138}$La summing internally with K$\alpha$ X-rays of 32 keV. Moreover, it is worth noting that the band of alpha particles observed in the spectrum, reported earlier in [3], has at least an order of one magnitude lower intensity, reflecting a significant improvement of the new LaBr$_3$:Ce and LaCl$_3$:Ce scintillating crystals.

3.2 LaBr$_3$:Ce and LaCl$_3$:Ce Energy Resolution

Energy resolution of LaBr$_3$:Ce and LaCl$_3$:Ce were compared with the resolution of CZT (currently used for nuclear safeguards) and NaI(Tl) [7]. We note that good spectral resolution is required for isotopic identification, especially if peaks are closely grouped together as seen in HEU and $^{239}$Pu, or for identification of isotopes in a mixed source. For example, $^{133}$Ba, has four main peaks grouped fairly close together (separated by just over 100 keV) shows the importance of adequate spectral resolution for isotopic identification.
Noted in Figure 3, the NaI(Tl) detector is unable to resolve all four key peaks of $^{133}\text{Ba}$. Whereas the CZT and LaBr$_3$:Ce and LaCl$_3$:Ce assembly are able to resolve these emission line groupings. Good spectral resolution and high efficiency becomes important when working with sources that have a complicated decay scheme. Similarly, $^{152}\text{Eu}$ has a complicated decay scheme and emissions over a large range of energy, with relatively close peaks at both ends of the energy range.

![Figure 3. $^{133}\text{Ba}$ Energy resolution comparison between LaBr$_3$:Ce and LaCl$_3$:Ce, NaI(Tl) and CdZnTe](image)

![Figure 4. $^{152}\text{Eu}$ energy spectrum comparison between LaBr$_3$:Ce, LaCl$_3$:Ce, NaI(Tl) and CdZnTe](image)
As seen in Figure 4, LaBr$_3$:Ce and LaCl$_3$:Ce were able to resolve closely spaced emission lines of $^{152}$Eu, comparing favourably to CZT. When a similar $^{152}$Eu spectrum was taken with NaI(Tl), none of the relatively close spectral lines could be resolved even at low to moderate energies where the detector resolution should have been at its comparable best as seen in Figure 8.

Figure 5 and 6 are included for the similar comparisons for $^{239}$Pu and HEU.

Figures 5. $^{239}$Pu Energy spectrum measured using LaBr$_3$:Ce, LaCl$_3$:Ce, NaI(Tl) and CZT.

Figures 6. Energy spectrum of HEU measured using LaBr$_3$:Ce, LaCl$_3$:Ce, NaI(Tl) and CZT.
Figure 7 illustrates differences in resolution between detectors by plotting the FWHM as a function of the gamma energies. We observe that FWHM of LaBr$_3$:Ce and LaCl$_3$:Ce is a good compromise between CZT and NaI(Tl) detectors. An improvement that LaBr$_3$:Ce and LaCl$_3$:Ce has over CZT, even though the FWHM is not as good, is that it has a Gaussian emission lines distribution in contrast to the asymmetry seen with CZT. Gaussian distribution is desirable for most peak analysis and identification process. In general these programs will use a form of peak fitting and 2$^{nd}$ order differential analysis as a process to interpret spectral information that may be hidden in electronic noise. Unique peak analysis and identification program [11] is used for asymmetric peaks.
Figure 8. Energy resolution (FWHM/E) comparisons of LaBr$_3$:Ce ●, LaCl$_3$:Ce ▲, NaI (Te) □ and CZT ×.

As seen by the energy resolution in Figure 8 (defined as FWHM/E) both LaBr$_3$:Ce and LaCl$_3$:Ce follow the resolution curve of CZT relatively closely and is much more comparable to CZT than NaI(Tl). The resolution of the LaBr$_3$:Ce and LaCl$_3$:Ce appears to be at its best for moderate to high energies 150-450 keV (region of interest to Safeguards application) and above. We note that there has been published evidence [1] that below these energies (approximately 100 keV) the resolution becomes comparable to NaI(Tl) (as seen in figure. 8) where the resolution of NaI(Tl) and LaBr$_3$:Ce and LaCl$_3$:Ce are similar.

For energies below 100 keV NaI(Tl) could be preferable over LaBr$_3$:Ce and LaCl$_3$:Ce. The resolution, seen in figure 8, of $^{137}$Cs 662 keV line for NaI(Tl) is ~ 6.7% compared with ~ 1.8% of CZT and ~ 3.7% of LaBr$_3$:Ce and LaCl$_3$:Ce. For the 59.5 keV emission from $^{241}$Am the resolution for NaI(Tl) is ~13.3% and for CZT the resolution ~7.9% while LaBr$_3$:Ce has a resolution of ~13.4%. This demonstrates that LaBr$_3$:Ce and LaCl$_3$:Ce compares favourably with CZT.

3.3 LaBr$_3$:Ce, LaCl$_3$:Ce, Efficiency Comparison

Efficiency between the different detector types was compared; the volume of scintillators LaBr$_3$:Ce(12.9 cm$^3$), LaCl$_3$:Ce (12.9 cm$^3$) and NaI(Tl) normalised but not to the CZT detector. The
CZT detector used in the comparison was a Ritek 500/s 'large volume' hemispherical detector (volume 0.5 cm$^3$). Efficiency comparisons were made directly between normalised scintillators and the CZT detector. CZT detectors larger than 0.2 cm$^3$ tend to exhibit charge trapping and consequently have poor spectral response and are used mostly as counters; to normalise the CZT volume to the scintillators would be counterproductive.

![Efficiency comparison graph](image)

**Figure 9.** Relative efficiency comparison between of LaBr$_3$:Ce o, LaCl$_3$:Ce ▲, NaI (Te) □, CZT x.

From Figure 9 we observe that LaBr$_3$:Ce and LaCl$_3$:Ce have relative efficiency greater than one magnitude when compared to CZT detector, and are similar in relative efficiency as the NaI(Tl) detector with twice the volume.

### 3.4 Comparison of Radium and naturally occurring Thorium.

Radium and thorium were used to determine high energy resolution by observing the individual spectra from each and then a combined spectrum of both mixed together. $^{226}$Ra and $^{232}$Th in equilibrium with their daughter products were used. This was done because the spectral lines from both are very similar to each other and to other radionuclides of interest. For both LaBr$_3$:Ce / LaCl$_3$:Ce, and NaI(Tl) measurements were taken for 3000 seconds. $^{226}$Ra and $^{232}$Th and then followed, similarly, with measurement of the mixed $^{226}$Ra and $^{232}$Th source.

The peaks used in identification [Nuclear Data ref.12] of $^{226}$Ra and $^{232}$Th are not necessarily from the intrinsic decay peaks of the parent nuclei, but are from the peaks of their shorter lived...
daughter products, which are in equilibrium with the parent. Peaks identified for $^{226}$Ra (in keV) are 186.11 ($^{226}$Ra), 241.91 ($^{214}$Pb), 351.86 ($^{214}$Pb and $^{214}$Bi), 609.31 ($^{214}$Bi), 1120.27 ($^{214}$Bi) and 1764.49 ($^{214}$Bi). Peaks identified for $^{232}$Th in keV are 238.57 ($^{212}$Pb), 583.02 ($^{208}$Tl), 727.24 ($^{212}$Bi, $^{212}$Bi, $^{228}$Ac and $^{212}$Pom), and 2614.35 ($^{212}$Pom and $^{208}$Tl). These peaks are the most intense peaks in the spectra, as can be seen in Figure 10.

![Energy spectrum comparison between Ra, Th and Ra + Th.](image)

Figures 10 shows that the presence of Radium and Thorium can be detected with LaBr$_3$:Ce and LaCl$_3$:Ce. However, several daughter products for $^{232}$Th are very close in energy and cannot be resolved; they overlap, and contribute to the intensity of a combined peak for a moderate resolution detector.

4. Summary

LaBr$_3$:Ce and LaCl$_3$:Ce appear to be an excellent alternative for NaI(Tl) and a possible replacement for CdZnTe in a variety of applications for gamma spectroscopy where moderate energy resolution and high detection efficiency is required. Also evident, for long counting times, are three intrinsic characteristic spectral lines (2 gamma and 1 X-ray) of the $^{138}$La which appear as a residual in spectrum display. However, being aware of these emission lines, gamma spectroscopy
using LaBr₃:Ce, LaCl₃:Ce should not present problem. LaBr₃:Ce and LaCl₃:Ce scintillators can be used in a wide range of applications, such as 4πβ-γ coincidence - counting, isotopic identification, and Safeguards applications for monitoring of nuclear materials.

Acknowledgments

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References

[10] Professor Marek Mozynski of the Soltan Institute for Nuclear Studies, private communication.
Chapter Eleven

Activity measurements of the radionuclide $^{153}$Sm for the ANSTO, Australia in the ongoing comparison BIPM.RI(II)-K1.Sm-153

G, Ratel*, C. Michotte*, M.I. Reinhard†, D. Alexiev†, L. Mo†

*Bureau International des Poids et Mesures, France.

†Australian Nuclear Science and Technology Organisation (ANSTO), New Illawarra Road, Lucas Heights 2234 NSW Australia

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Author’s contribution

G, Ratel
Carried out comparison experiment and data analysis, wrote manuscript.

C. Michotte
Carried out comparison experiment and data analysis, contributed to manuscript writing.

M.I. Reinhard
Performed primary and secondary standardisation of $^{153}$Sm.

D. Alexiev
Contributed to manuscript editing.

L. Mo (candidate)
Organised the international comparison, performed secondary standardization measurement and impurity analysis for the comparison samples.
Activity measurements of the radionuclide $^{152}\text{Sm}$ for the ANSTO, Australia in the ongoing comparison BIPM RI(II)-K1 Sm-153

G. Ratei*, C. Michotte*, D. Alexiev§, Li Mo§
*BIPM, §ANSTO, Australia

Abstract

In 2004, the Australian Nuclear Science and Technology Organisation (ANSTO) submitted two samples of known activity of $^{152}\text{Sm}$ to the International Reference System (SIR). The value of the activity submitted was about 920 MBq. This key comparison result has been added to the matrix of degrees of equivalence in the key comparison database that now contains five results, identifier BIPM RI(II)-K1 Sm-153.

1. Introduction

The SIR for activity measurements of $\gamma$-ray-emitting radionuclides was established in 1976. Each national metrology institute (NMI) may request a standard ampoule from the BIPM that is then filled (3.6 g) with the radionuclide in liquid (or gaseous) form. The NMI completes a submission form that details the standardization method used to determine the absolute activity of the radionuclide and the full uncertainty budget for the evaluation. The ampoules are sent to the BIPM where they are compared with standard sources of $^{226}\text{Ra}$ using pressurized ionization chambers. Details of the SIR method, experimental set-up and the determination of the equivalent activity, $A_e$, are all given in [1].

From its inception until 31 December 2004, the SIR has measured 872 ampoules to give 634 independent results for 62 different radionuclides. The SIR makes it possible for national laboratories to check the reliability of their activity measurements at any time. This is achieved by the determination of the equivalent activity of the radionuclide and by comparison of the result with the key comparison reference value determined from the results of primary realizations. These comparisons are described as BIPM ongoing comparisons and the results form the basis of the BIPM key comparison database (KCDB) that was set up under the CIPM Mutual Recognition Arrangement (MRA) [2]. The comparison described in this report is known as the BIPM RI(II)-K1 Sm-153 key comparison.

2. Participants

In addition to the ANSTO, four NMIs have submitted ampoules for the comparison of $^{152}\text{Sm}$ activity measurements since 1998. The ANSTO details are given in Table 1 and the details of the other participants are given in [3].
Table 1. Details of the ANSTO participation in the comparison BIPM.RI(II)-K1.Sm-153

<table>
<thead>
<tr>
<th>NMI</th>
<th>Full name</th>
<th>Country</th>
<th>Regional metrology organization</th>
<th>Date of measurement at the BIPM</th>
</tr>
</thead>
<tbody>
<tr>
<td>ANSTO</td>
<td>Australian Nuclear Science and Technology Organisation</td>
<td>Australia</td>
<td>APMP</td>
<td>2004-09-22</td>
</tr>
</tbody>
</table>

3. NMI standardization methods

Each NMI that submits ampoules to the SIR has measured the activity either by a primary standardization method or by using a secondary method, for example a calibrated ionization chamber. In the latter case, the traceability of the calibration needs to be clearly identified to ensure that any correlations are taken into account.

A brief description of the standardization methods for the laboratory, the activities submitted and the relative standard uncertainties ($k = 1$) are given in Table 2. The uncertainty budget is given in Appendix 1. The acronyms used for the measurement methods are given in Appendix 2.

Table 2. Standardization methods of the ANSTO for $^{153}$Sm

<table>
<thead>
<tr>
<th>NMI</th>
<th>Method used and acronym (see Appendix 2)</th>
<th>Half-life / d</th>
<th>Activity / kBq</th>
<th>Reference date</th>
<th>Relative standard uncertainty $\times 100$ by method of evaluation</th>
</tr>
</thead>
<tbody>
<tr>
<td>ANSTO</td>
<td>Pressurized ionization chamber calibrated by $4\pi\beta-\gamma$ coincidence meas. in June 2003 $4\pi-???BP-NA-GR-CO$</td>
<td>1.928 49 (11) [4]</td>
<td>915 900 907 600*</td>
<td>04-09-07 12 h UTC</td>
<td>0.03 0.94</td>
</tr>
</tbody>
</table>

* two ampoules submitted

The half-life used by the BIPM is 46.284 (4) hours [3] which is in very good agreement with the more recent evaluation by the same author published in [4].

Details regarding the solution submitted are shown in Table 3, including any impurities, when present, as identified by the laboratory. Recently the BIPM has developed a standard method for evaluating the activity of impurities using a calibrated Ge(Li) spectrometer [5]. The CCRI(II) agreed in 1999 [7] that this method should be followed according to the protocol described in [8] when an NMI makes such a request or when there appear to be discrepancies.
For this submission, the measurement of the ampoule at the BIPM was delayed by a week due to transport/logistics problems so that the contribution of the impurities to the ionization current became important. A 5% discrepancy in the SIR result when compared with the KCRV was observed and this motivated a γ-spectrometry measurement at the BIPM. The resulting relative activities of $^{152}$Eu, $^{154}$Eu and $^{155}$Eu are in agreement with the ANSTO values within one or two combined standard uncertainties. In addition, a $^{156}$Eu impurity was measured at the BIPM, as given in Table 3. The consequences on the ANSTO SIR results are discussed in section 4.

Table 3. Details of the solution of $^{152}$Sm submitted

<table>
<thead>
<tr>
<th>NMI</th>
<th>Chemical composition</th>
<th>Solvent conc. / (mol dm$^{-3}$)</th>
<th>Carrier conc. / (µg g$^{-1}$)</th>
<th>Density / (g cm$^{-3}$)</th>
<th>Relative activity of any impurity†</th>
</tr>
</thead>
</table>
| ANSTO | Sm in HCl            | 0.1                             | Sm: 720                       | 1.04                   | $^{152}$Eu: $1.20 \times 10^{-2}$% * $^{152}$Eu: $1.43 \times 10^{-3}$% $^{154}$Eu: $7.8 \times 10^{-4}$% $^{155}$Eu: $2.3 \times 10^{-4}$% $^{156}$Eu: $1.16 \times 10^{-3}$% *

1 the ratio of the activity of the impurity to the activity of $^{152}$Sm at the reference date
2 negligible influence on the SIR measurement
3 measured at the BIPM

4. Results

All the submissions to the SIR since its inception in 1976 are maintained in a database known as the "mother-file". The previous activity measurements for $^{152}$Sm arise from seven ampoules and the SIR equivalent activity, $A_{eq}$, for each ampoule is given in [3] for each NMI, i. The SIR equivalent activities for the ANSTO ampoule are given in Table 4. The date of measurement in the SIR is given in Table 1 and is used in the KCDB and all references in this report. The relative standard uncertainty arising from the measurements in the SIR is also shown. This uncertainty is additional to that declared by the NMI for the activity measurement shown in Table 2. Although activities submitted are compared with a given source of $^{226}$Ra, all the SIR results are normalized to the radium source number 5 [1].

The ionization current measurement by the ANSTO at the reference date was insensitive to the europium impurities as the ionization current was then dominated by the $^{152}$Sm activity. Unfortunately, at the time of the SIR measurement almost 15 days later, the correction for impurities is 1.187 and becomes 1.234 when the $^{156}$Eu impurity detected at the BIPM is also taken into account. The final result corrected for all impurities in Table 4 is in agreement with the KCRV within the combined standard uncertainty.

Measurements repeated at the BIPM one day later produced comparison results in agreement within less than $2 \times 10^{-3}$.
Table 4. Results of SIR measurements of $^{153}$Sm for the ANSTO

<table>
<thead>
<tr>
<th>NMI</th>
<th>Mass of solution / g</th>
<th>Activity submitted/ kBq</th>
<th>N° of Ra source used</th>
<th>SIR $A_e$ / MBq</th>
<th>Relative uncertainty from SIR</th>
<th>Total uncertainty $u_{c1}$ / MBq</th>
</tr>
</thead>
<tbody>
<tr>
<td>ANSTO</td>
<td>3.6353</td>
<td>915 900</td>
<td>1</td>
<td>565.6</td>
<td>1.1 x 10^{-3} *</td>
<td>8.0</td>
</tr>
<tr>
<td></td>
<td>3.6024</td>
<td>907 600</td>
<td></td>
<td>565.3 *</td>
<td></td>
<td>8.1</td>
</tr>
</tbody>
</table>

*The mean of the $A_e$ values is used with an averaged uncertainty, as attributed to an individual entry [9]
*corrected for ANSTO impurities and the $^{153}$Eu impurity measured at the BIPM
* due to the combination of a long delay between the reference date and the SIR measurement, and the uncertainties of the activity of the impurities.

4.1 The key comparison reference value

The key comparison reference value for $^{153}$Sm is 573 700 (1 200) kBq as given in [3]. Although the KCRV may be modified whenever an NMI participates, such modifications are only made by the CCRI(II), normally during one of its biennial meetings. However, due to the uncertainty coming from the large impurity correction of the ANSTO SIR measurement, it is not recommended to include this result in the KCRV.

4.2 Degrees of equivalence

Every NMI that has submitted ampoules to the SIR is entitled to have one result included in Appendix B of the KCDB as long as the NMI is a signatory or designated institute listed in the MRA. Normally, the most recent result is the one included. Any NMI may withdraw its result only if all the participants agree.

The degree of equivalence of a given measurement standard is the degree to which this standard is consistent with the key comparison reference value [2]. The degree of equivalence is expressed quantitatively in terms of the deviation from the key comparison reference value and the expanded uncertainty of this deviation ($k=2$). The degree of equivalence between any pair of national measurement standards is expressed in terms of their difference and the expanded uncertainty of this difference and is independent of the choice of key comparison reference value.

4.2.1 Comparison of a given NMI with the KCRV

The degree of equivalence of a particular NMI, $i$, with the key comparison reference value is expressed as the difference between the results

$$D_i = A_{ei} - \text{KCRV}$$

and the expanded uncertainty ($k=2$) of this difference, $U_i$, known as the equivalence uncertainty, hence

$$U_i = 2u_{l_i},$$

taking correlations into account as appropriate [8].
4.2.2 *Comparison of any two NMIs with each other*

The degree of equivalence, $D_{ij}$, between any pair of NMIs, $i$ and $j$, is expressed as the difference in their results

$$D_{ij} = D_i - D_j = A_i - A_j$$  \hspace{1cm} (3)

and the expanded uncertainty of this difference $U_{ij}$ where

$$u_{ij}^2 = u_i^2 + u_j^2 - 2u(A_i, A_j)$$  \hspace{1cm} (4)

where any obvious correlations between the NMIs (such as a traceable calibration) are subtracted using the covariance $u(A_i, A_j)$, as are normally those correlations coming from the SIR.

The uncertainties of the differences between the values assigned by individual NMIs and the key comparison reference value (KCRV) are not necessarily the same uncertainties that enter into the calculation of the uncertainties in the degrees of equivalence between a pair of participants. Consequently, the uncertainties in the table of degrees of equivalence cannot be generated from the column in the table that gives the uncertainty of each participant with respect to the KCRV. However, the effects of correlations have been treated in a simplified way as the degree of confidence in the uncertainties themselves does not warrant a more rigorous approach.

Table 5 shows the matrix of all the degrees of equivalence as they will appear in Appendix B of the KCDB. It should be noted that for consistency within the KCDB, a simplified level of nomenclature is used with $A_{ij}$ replaced by $x_i$. The introductory text is that agreed for the comparison. The graph of the first column of results in Table 5, corresponding to the degrees of equivalence with respect to the KCRV (identified as $x_k$ in the KCDB), is shown in Figure 1. The graphical representation indicates in part the degree of equivalence between the NMIs but does not take into account the correlations between the different NMIs. However, the matrix of degrees of equivalence shown in yellow in Table 5 does take the known correlations into account.

**Conclusion**

The result for the ANSTO in this ongoing comparison agrees with the KCRV within the combined standard uncertainties.

The BIPM ongoing key comparison for $^{155}$Sm, BIPM RI(II)-K1.Sm-153 currently comprises five results. These have been analysed with respect to the KCRV determined for this radionuclide, and with respect to each other. The matrix of degrees of equivalence has been approved by the CCRI(II) and is published in the BIPM key comparison database.

Other results may be added as and when NMIs contribute $^{152}$Sm activity measurements to this comparison or take part in linked comparisons.
Acknowledgements

The authors would like to thank Justin Davies for the measurements at the ANSTO, Sammy Courte of the BIPM for his measurement in the SIR, and Dr P.J. Allisy-Roberts of the BIPM for editorial assistance.

References


Table 5. Table of degrees of equivalence and introductory text for $^{153}$Sm

Key comparison BIPM.RI(II)-K1.Sm-153

MEASURAND: Equivalent activity of $^{153}$Sm

Key comparison reference value: the SIR reference value $x_c$ for this radionuclide is 573.7 MBq, with a standard uncertainty, $u_c = 1.2$ MBq (see Section 4.1 of the Final Report). The value $x_i$ is taken as the equivalent activity for laboratory $i$.

The degree of equivalence of each laboratory with respect to the reference value is given by a pair of terms: $D_i = (x_i - x_c)$ and $U_i$, its expanded uncertainty ($k = 2$), both expressed in MBq, and $U_i = 2[(1 - 2/n)u_i^2 + (1/n^2)\sum u_{ij}^2]^{1/2}$ when each laboratory has contributed to the calculation of $x_c$.

The degree of equivalence between two laboratories is given by a pair of numbers: $D_y = D_i - D_j = (x_i - x_j)$ and $U_y$ its expanded uncertainty ($k = 2$), both expressed in MBq. The approximation $U_y^2 = D_y^2 + U_i^2 + U_j^2$ is used in the following table.

<table>
<thead>
<tr>
<th>Lab $i$</th>
<th>NIST</th>
<th>PTB</th>
<th>NPL</th>
<th>BNM-LNHB</th>
<th>ANSTO</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$D_i$</td>
<td>$U_i$</td>
<td>$D_y$</td>
<td>$U_y$</td>
<td>$D_y$</td>
</tr>
<tr>
<td>NIST</td>
<td>3.1</td>
<td>7.4</td>
<td>5.1</td>
<td>11.3</td>
<td>2.1</td>
</tr>
<tr>
<td>PTB</td>
<td>-2.0</td>
<td>5.4</td>
<td>-5.1</td>
<td>11.3</td>
<td>-3.0</td>
</tr>
<tr>
<td>NPL</td>
<td>1.0</td>
<td>5.2</td>
<td>-2.1</td>
<td>11.0</td>
<td>3.0</td>
</tr>
<tr>
<td>BNM-LNHB</td>
<td>-2.0</td>
<td>3.6</td>
<td>-5.1</td>
<td>9.7</td>
<td>0.0</td>
</tr>
<tr>
<td>ANSTO</td>
<td>-8.2</td>
<td>16.5</td>
<td>-11.3</td>
<td>18.7</td>
<td>-6.2</td>
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</table>
Figure 1. Graph of degrees of equivalence with the KCRV for $^{153}$Sm
(as it appears in Appendix B of the MRA)
Appendix 1. Uncertainty budgets for the activity of $^{153}$Sm submitted to the SIR

**Uncertainty budget for the ANSTO measurement (2004)**

<table>
<thead>
<tr>
<th>Contributions due to</th>
<th>$u_f \times 10^4$</th>
<th>evaluated by method</th>
</tr>
</thead>
<tbody>
<tr>
<td>Current measurement</td>
<td>3.1</td>
<td>–</td>
</tr>
<tr>
<td>Radium stability correction</td>
<td>–</td>
<td>20.0</td>
</tr>
<tr>
<td>$^{153}$Sm calibration factor</td>
<td>–</td>
<td>92.0</td>
</tr>
<tr>
<td>Weighing</td>
<td>–</td>
<td>0.2</td>
</tr>
<tr>
<td>Background current</td>
<td>0.0</td>
<td>–</td>
</tr>
<tr>
<td>$^{153}$Sm decay correction</td>
<td>–</td>
<td>2.0</td>
</tr>
<tr>
<td>$^{155}$Eu impurity correction</td>
<td>–</td>
<td>1.1</td>
</tr>
<tr>
<td>$^{155}$Eu impurity correction</td>
<td>–</td>
<td>0.2</td>
</tr>
<tr>
<td>$^{154}$Eu impurity correction</td>
<td>–</td>
<td>0.1</td>
</tr>
<tr>
<td>$^{155}$Eu impurity correction</td>
<td>–</td>
<td>0.0</td>
</tr>
<tr>
<td>Quadratic summation</td>
<td>3.1</td>
<td>94.1</td>
</tr>
<tr>
<td>Relative combined standard uncertainty, $u_c$</td>
<td>94.2</td>
<td></td>
</tr>
</tbody>
</table>
Appendix 2. Acronyms used to identify different measurement methods

Each acronym has six components: geometry, detector (1), radiation (1), detector (2), radiation (2), mode. When a component is unknown, ?? is used and when it is not applicable 00 is used.

<table>
<thead>
<tr>
<th>Geometry</th>
<th>acronym</th>
<th>Detector</th>
<th>acronym</th>
</tr>
</thead>
<tbody>
<tr>
<td>4π</td>
<td>4P</td>
<td>proportional counter</td>
<td>PC</td>
</tr>
<tr>
<td>defined solid angle</td>
<td>SA</td>
<td>press. prop counter</td>
<td>PP</td>
</tr>
<tr>
<td>2π</td>
<td>2P</td>
<td>liquid scintillation counting</td>
<td>LS</td>
</tr>
<tr>
<td>undefined solid angle</td>
<td>UA</td>
<td>NaI(Tl)</td>
<td>NA</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Ge(HP)</td>
<td>GH</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Ge(LI)</td>
<td>GL</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Si(LI)</td>
<td>SL</td>
</tr>
<tr>
<td></td>
<td></td>
<td>CsI(Tl)</td>
<td>CS</td>
</tr>
<tr>
<td></td>
<td></td>
<td>ionization chamber</td>
<td>IC</td>
</tr>
<tr>
<td></td>
<td></td>
<td>grid ionization chamber</td>
<td>GC</td>
</tr>
<tr>
<td></td>
<td></td>
<td>bolometer</td>
<td>BO</td>
</tr>
<tr>
<td></td>
<td></td>
<td>calorimeter</td>
<td>CA</td>
</tr>
<tr>
<td></td>
<td></td>
<td>PIPS detector</td>
<td>PS</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Radiation</th>
<th>acronym</th>
<th>Mode</th>
<th>acronym</th>
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</thead>
<tbody>
<tr>
<td>positron</td>
<td>PO</td>
<td>efficiency tracing</td>
<td>ET</td>
</tr>
<tr>
<td>beta particle</td>
<td>BP</td>
<td>internal gas counting</td>
<td>IG</td>
</tr>
<tr>
<td>Auger electron</td>
<td>AE</td>
<td>CIEMAT/NIST</td>
<td>CN</td>
</tr>
<tr>
<td>conversion electron</td>
<td>CE</td>
<td>sum counting</td>
<td>SC</td>
</tr>
<tr>
<td>mixed electrons</td>
<td>ME</td>
<td>coincidence</td>
<td>CO</td>
</tr>
<tr>
<td>bremsstrahlung</td>
<td>BS</td>
<td>anti-coincidence</td>
<td>AC</td>
</tr>
<tr>
<td>gamma rays</td>
<td>GR</td>
<td>coincidence counting</td>
<td>CT</td>
</tr>
<tr>
<td>X-rays</td>
<td>XR</td>
<td>anti-coincidence with efficiency tracing</td>
<td>AT</td>
</tr>
<tr>
<td>photons (X + γ)</td>
<td>PH</td>
<td>triple-to-double coincidence ratio counting</td>
<td>TD</td>
</tr>
<tr>
<td>photons + electrons</td>
<td>PE</td>
<td>selective sampling</td>
<td>SS</td>
</tr>
<tr>
<td>alpha - particle</td>
<td>AP</td>
<td>high efficiency</td>
<td>HE</td>
</tr>
<tr>
<td>mixture of various radiation</td>
<td>MX</td>
<td>digital coincidence counting</td>
<td>DC</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Examples</th>
<th>method</th>
<th>acronym</th>
</tr>
</thead>
<tbody>
<tr>
<td>4π(PO)β→γ coincidence counting</td>
<td>4P-PC-β-NA-GR-00-00</td>
<td></td>
</tr>
<tr>
<td>4π(PO)β→γ coincidence counting eff. trac.</td>
<td>4P-PC-β-NA-GR-CT</td>
<td></td>
</tr>
<tr>
<td>defined solid angle α-particle counting with a PIPS detector</td>
<td>SA-PS-AP-00-00-00</td>
<td></td>
</tr>
<tr>
<td>4π(PO)X-γ(0e-HP) coincidence counting</td>
<td>4P-PO-MX-γ-GR-AC</td>
<td></td>
</tr>
<tr>
<td>4π Cs-137 AX γ counting</td>
<td>4P-CSAP-00-00-00</td>
<td></td>
</tr>
<tr>
<td>calibrated IC</td>
<td>4P-IC-GR-00-00-00</td>
<td></td>
</tr>
<tr>
<td>internal gas counting</td>
<td>4P-PC-BP-00-00-10</td>
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</tbody>
</table>
Chapter Twelve

Conclusion

This study showed solutions of various problems encountered in standardising pure $\beta$ and $\beta-\gamma$ emitting radionuclides in various physical forms. The conclusions obtained from the study can be summarised below:

1. For the absolute activity measurement of pure $\beta$-emitters in liquid form using the liquid scintillation TDCR and CIEMAT/NIST method, the basic requirement of the system set-up is to set the discrimination level just before the single photoelectron peak so that all single photoelectron pulses caused by disintegration are counted. The study showed that this was particularly critical for low energy beta emitters such as $^3$H and $^{63}$Ni. The error in activity, when the requirement was difficult to meet, could be eliminated by applying a fraction-correction to the efficiency calculation. The fraction-correction corrected the proportion of the single photoelectron peak being rejected.

2. A new approach developed in this study for the standardisation of $^{90}$Y microspheres was to apply a highly quantitative chemical-digestion technique in the standardisation procedure. In this way, the activity standard for $^{90}$Y solution, developed using the liquid scintillation TDCR and CIEMAT/NIST methods, was transferred to microspheres.

3. A highly accurate method was investigated in this study for the absolute activity determination of $^{198}$Au in the form of Au wire, Al-Au (0.112% of Au) alloy wire and Au foil, using a $4\pi\beta-\gamma$ coincidence-counting technique. Monte Carlo simulation technique was used for the determination of the decay-scheme-dependent correction for the coincidence equation.

4. To improve the energy resolution, without losing detection efficiency for a photon detector used in $4\pi\beta-\gamma$ coincidence counting, a study was undertaken to show that new scintillators such as LaBr$_3$:Ce and LaCl$_3$:Ce, activated with cerium (Ce$^{3+}$) atoms as the primary scintillation source, offer an excellent alternative to the NaI scintillator which has the disadvantage of low energy resolution.

5. The activity standards developed in this work have been successfully used in the neutron flux measurements for the hot commissioning process of the OPAL reactor (ANSTO) and the calibration of ionisation chambers used by the Australian radiopharmaceutical producers and hospitals.

TDCR and CIEMAT/NIST methods are the preferred methods for the absolute activity measurement of pure $\beta$-emitters and electron capture nuclides in a liquid form. They have a stringent requirement that the experiment condition meets the efficiency calculation model. The
study of the single photoelectron peak carried out in this thesis can be extended to electron capture nuclides. When using the TDCR and CIEMAT/NIST methods to measure solid sources (such as $^{80}$Y microspheres), a destructive method has to be used to dissolve the solid substance.

4π$\beta-\gamma$ coincidence counting technique for the absolute activity measurement of $\beta-\gamma$ emitters requires no separate measurement of detection efficiency. But a counting source has to be very thin in order to maximise the beta counting efficiency. However for a thick source with a complex decay scheme (such as $^{198}$Au wires and foils), a decay-scheme-dependent correction has to be applied to the coincidence equation to correct for those unwanted radiations. Monte Carlo simulation provides a great resolution for the determination of the correction. It is a non-destructive method. There is limitation with Monte Carlo modelling of electrons transporting through thin foils, as noted from the results of MCNP simulation of $^{198}$Au foils. The problem resides in optimising the multiple scattering algorithm to deal with material boundaries and backscattering rather than the angular distributions in the bulk. Further investigation in modelling thin foils may be carried out in the future using a more modern and suitable simulation tool such as GEANT4. It will allow a greater flexibility of modelling the experimental set-up and the physics configuration of the simulation.
Appendix

List of other publications and conference presentations


