

STANDARDIZATION OF ^{111}In

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ABSTRACT

The radionuclide ^{111}In is widely used in medical, in vivo diagnosis and research. It is therefore necessary to know the specific activity of the ^{111}In sources so that the proper dose may be administered to give the proper results without endangering the patient. In the past, this has only been done by an indirect method. Certified sources of ^{111}In are obtained from the National Bureau of Standards, Radioactivity Group, and these are used to calibrate a counter. Subsequent samples are counted in the same manner to obtain the activity of the ^{111}In source.

This paper deals with the absolute standardization of any ^{111}In source in a well-type NaI(Tl) detector. The method does not require any certified source. However, a source of ^{111}In was calibrated by this method and by the NBS method to prove the reliability of the method. The results agreed within $\pm 2.0\%$.

This new method measures the coincidences between the two cascading gamma rays from the decay of ^{111}In ; 172 keV and 246 keV. The sample is measured through an absorber thick enough to absorb the Cd x-rays which interfere because they are also coincident with the gamma rays. Four values are required: 1) counts of 172 keV gamma ray alone; 2) counts of 246 keV gamma ray alone; 3) counts of coincidences between 172 and 246 keV gamma rays and 4) the total counts of the source. Using the equation developed by Brinkman, et al², the absolute disintegration rate can be calculated for any ^{111}In source.

INTRODUCTION

The specific activity of ^{111}In is presently determined at the National Bureau of Standards in the NBS "4 π " gamma pressure ionization chamber calibrated by 4 π (A,x) - γ coincidence efficiency-extrapolation

tion technique¹. This is very special equipment which is not readily available in most nuclear medicine, clinical or biomedical research laboratories which are using ¹¹¹In. Most commercial suppliers of ¹¹¹In have counters which are calibrated routinely through standards supplied by the NBS. These calibrated counters are used to calibrate sources which are supplied to customers.

The short half-life of ¹¹¹In (2.805 days) makes it impractical to keep a calibrated source in the laboratory. Also, calibrated instruments can change between calibrations with no way of knowing of the change until the next calibration. For these reasons, it seemed desirable to provide a method of standardizing ¹¹¹In sources in a laboratory using an instrument which is readily available in most laboratories: a well-type NaI(Tl) solid crystal scintillation system.

Brinkman, et al²⁻⁵, have developed the theory of the absolute standardization by the coincidence method for radionuclides which decay by emission of two coincident but different energy gamma rays. This is different from the decay by emission of two coincident gamma rays of the same energy⁶. Brinkman, et al developed the equation:

$$S = T + \frac{A_1 A_2}{A_{12}}$$

where: S is the sample disintegration rate, dpm,

T is the total count rate of ¹¹¹In sample in the counter,

A₁ is the count rate of one of the two coincident gamma rays when detected alone,

A₂ is the count rate of the other coincident gamma ray when detected alone,

and A₁₂ is the count rate of the coincident detection of the two gamma rays.

Measurement of the four rates: T, A₁, A₂ and A₁₂, would enable the calculation of the disintegration rate of any ¹¹¹In source. All of these rates can be obtained with a well-type NaI(Tl) solid crystal scintillation system. Using a multichannel analyzer to store the complete spectrum will allow for detailed analysis of the spectrum.

However, measurement over each of the appropriate energy ranges, one at a time or in groups, will also give the necessary data to calculate the source disintegration rate. This technique works only if all parts of the source are detected with the same efficiency. As with similar types of radionuclide calibration, high count-rates should be avoided.

NBS CALIBRATION

A stock solution of ^{111}In was prepared. Approximately 5 grams of the solution were placed in each of four glass vials provided by NBS. The exact weight was recorded and the vials were sealed by flame. Two of these vials were sent to the Radioactivity Group, NBS, for calibration. Two were kept at Beckman for future checks. At 0900 PST November 26, 1980, the two vials were calibrated. The average radioactivity concentration was $6.378 \times 10^8 \text{ m}^{-1}\text{g}^{-1}$ ($1.063 \times 10^7 \text{ s}^{-1}\text{g}^{-1}$) with a total uncertainty of $\pm 1.38\%$. A small impurity of $^{114\text{m}}\text{In}$ was also detected at an activity ratio for $^{114\text{m}}\text{In}/^{111}\text{In}$ of 4.3×10^{-4} ($\pm 20\%$). This impurity level will be treated in detail later.

^{111}In MEASUREMENT

The ^{111}In spectra were measured in a thru-well NaI(Tl) solid crystal scintillation detector connected to a multichannel analyzer (Nuclear Data ND-1100). Weighed aliquots of the stock ^{111}In solution were diluted and weighed aliquots of the dilution were measured. Figure 1 shows the spectrum of a ^{111}In sample (bare source). The spectrum shows the 172 keV peak, 195 keV (172 + 23 keV Cd K-x-ray), 247 keV, 270 keV (247 + 23 keV Cd K-x-ray) and 419 keV (and the unresolved 442 keV (419 + 23 keV Cd K-x-ray) and 465 (419 + two 23 keV Cd K-x-rays). Also seen at the low channels are the 23 keV Cd K-x-ray, 46 keV (two Cd K-x-rays) and Pb K-x-rays (from photoelectric effect on Pb shielding around the detector). Table 1 shows the results for calculation of the ^{111}In specific activity from this data. There is some uncertainty because the two gamma ray groups are not well separated. Also, the X-ray peaks at 23 keV (Cd K-x-ray), 46 keV (two Cd K-x-rays) and 75 keV (Pb K-x-ray from photoelectric effect of gamma rays on Pb shielding around the detector) are present and have to be

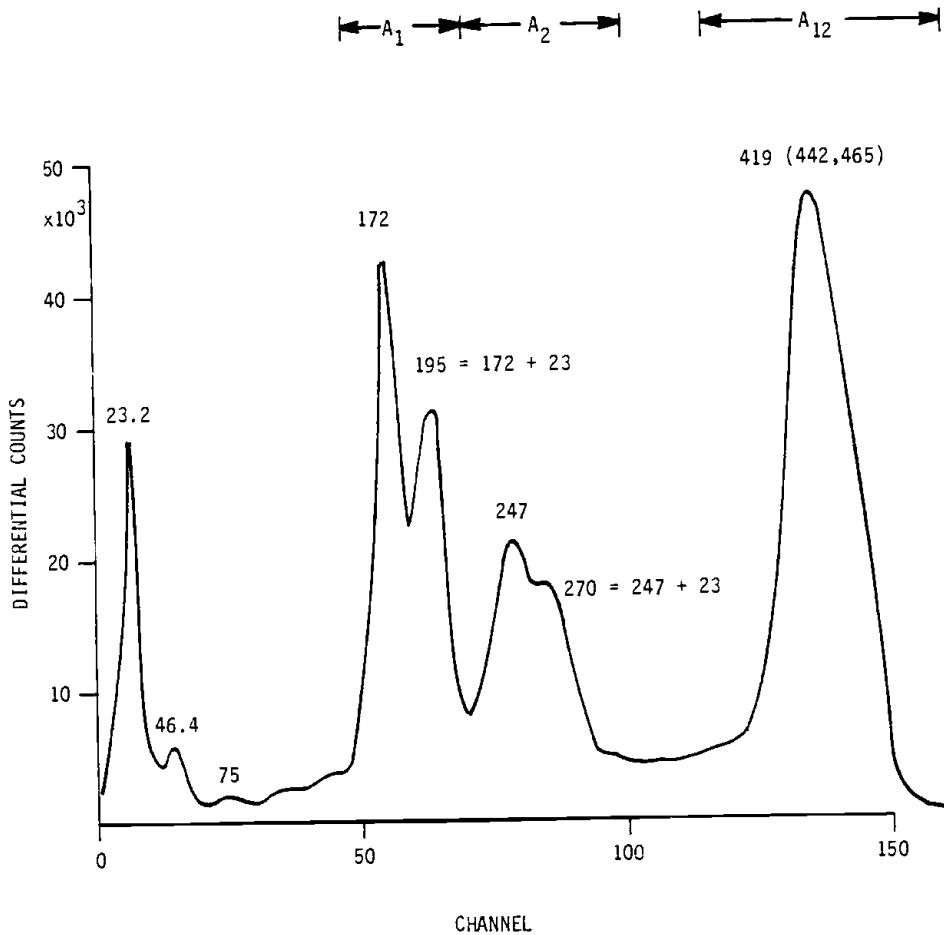


Figure 1. Pulse Height Spectrum of ^{111}In in Well-Thru NaI (Tl) Crystal Scintillation Detector. (No Absorber Around ^{111}In Source).

Table 1. Specific Activity of ^{111}In Source Under Varying Conditions

CONDITION	PST TIME	DATE	WEIGHT, GM	dpm/gm CORRECTED TO PST 0900 11-26-80
NBS	0900	11-26-80	5.24480	6.378×10^8 $\pm (1.38\%)$
Bare Source	1430	11-26-80	1.035×10^{-3}	6.468×10^8 (a) $\pm (3.00\%)$
Brass Sleeve Absorber				
Inside Well	1440	11-26-80	1.035×10^{-3}	6.377×10^8 (a) $\pm (2.00\%)$
Inside Well	1447	11-26-80	1.035×10^{-3}	6.385×10^8 (a) $\pm (2.00\%)$
No Pb Around Detector	1331	12-2-80	1.035×10^{-3}	6.394×10^8 (b) $\pm (2.00\%)$
Outside of Detector	1130	12-1-80	1.035×10^{-3}	6.410×10^8 (a) $\pm (3.00\%)$

(a) Corrected for Pb K- and L-x-ray peaks contribution to total count rate, T.

(b) No X-ray correction is required.

subtracted from the total count, T.

To eliminate some of these problems, a brass sleeve is inserted into the crystal well surrounding the ^{111}In sample. The brass was thick enough to absorb all of the 23 keV Cd K-x-rays, but only a minimum of the 172 and 247 keV gamma rays. This small loss does not alter the method of determining the disintegration rate of the ^{111}In with the brass sleeve. Note the double peaks have disappeared and the 172 and 247 peaks are easily resolved. Also, the 23 keV and 46 keV Cd K-x-ray peaks have disappeared. However, still present are the Pb K-x-ray (75 keV) and L-x-ray (12 keV) due to the photoelectric effect on the Pb shielding surrounding the detector. After correcting for

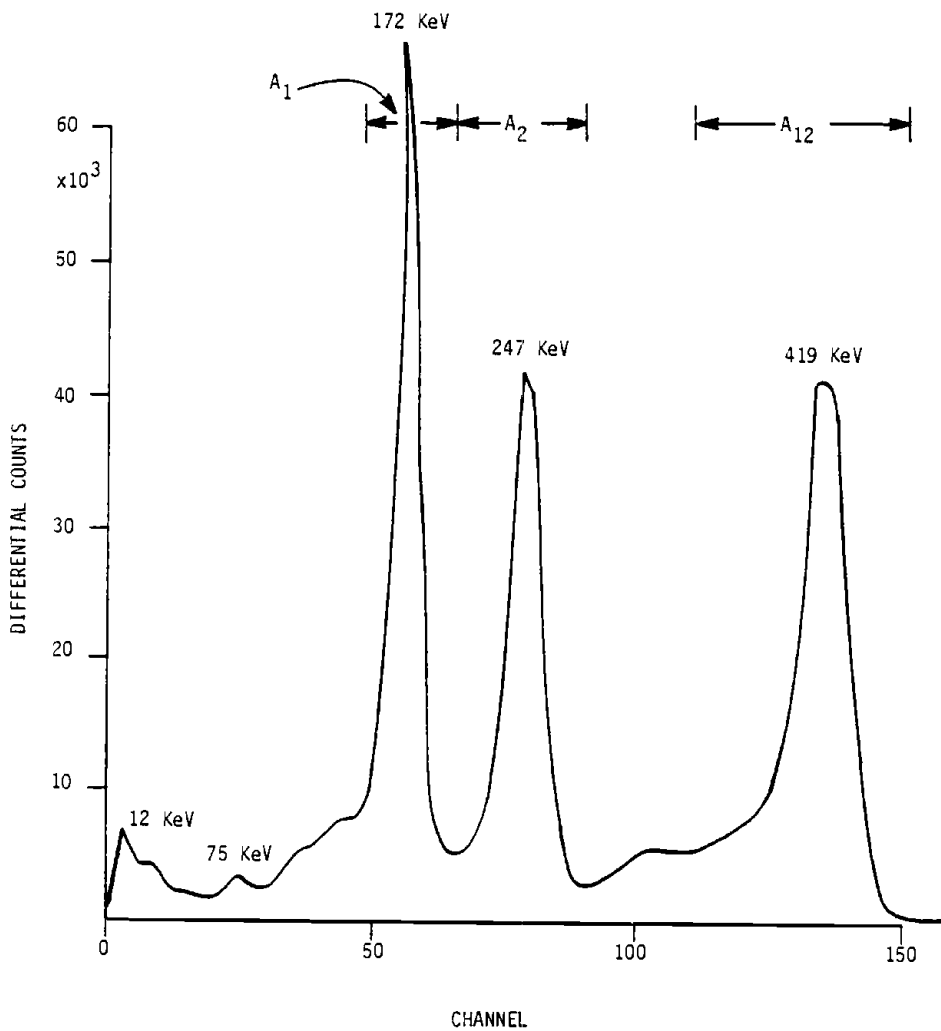


Figure 2. Pulse Height Spectrum of ^{111}In in Well-Thru NaI (Tl) Crystal Scintillation Detector with 0.3 cm-thick Brass Sleeve around the ^{111}In Source.

the Pb x-rays contribution to the value of T, the specific activity of the ^{111}In source was calculated and was in agreement with the NBS value (Table 1).

Figure 3 shows the ^{111}In spectrum (with brass sleeve) but all the surrounding Pb shield removed. The low energy peaks (75 and 12 keV) are missing. The ^{113}In specific activity calculated from this data agrees with the NBS value (see Table 1). This procedure will, of course, have much higher background corrections and may produce significant errors for low count rate samples. Also, other sources in the environment could cause serious errors.

Moving the ^{111}In source outside the detector greatly reduces the geometry and chance of coincidence, but the method is still applicable as long as the source is a point (as opposed to an extended or distributed source). Figure 4 shows the spectrum for an external point source. The specific activity calculated from this data (Table 1) agrees within 1% of the NBS value, but, of course, is more sensitive to errors due to the very low yield of coincidence events (i.e., dividing by a small number).

IMPURITY EFFECT

Because of the short half-life of ^{111}In (2.805 days), any impurities will have serious effects if they have considerably longer half-lives. The very small amount of $^{114\text{m}}\text{In}$ impurity reported by the NBS analysis was a specific activity ratio $^{114\text{m}}\text{In}/^{111}\text{In}$ at reference time of 4.3×10^{-4} ($\pm 20\%$). The half-life of $^{114\text{m}}\text{In}$ is 50 days. The activity ratio will increase more than 100-fold in just 20 days, which will introduce errors in this calculation method. In fact, subsequent calibrations, as given in Table 2, showed a high value of the ^{111}In specific activity which are caused by the increase in $^{114\text{m}}\text{In}$ activity relative to ^{111}In activity. However, this did produce between 1.21% and 2.34% increase in the calculated ^{111}In specific activity after 12 days.

CONCLUSION

This method of calibration of ^{111}In sources should make it much easier to ensure uses of proper amounts of ^{111}In . The method is easy

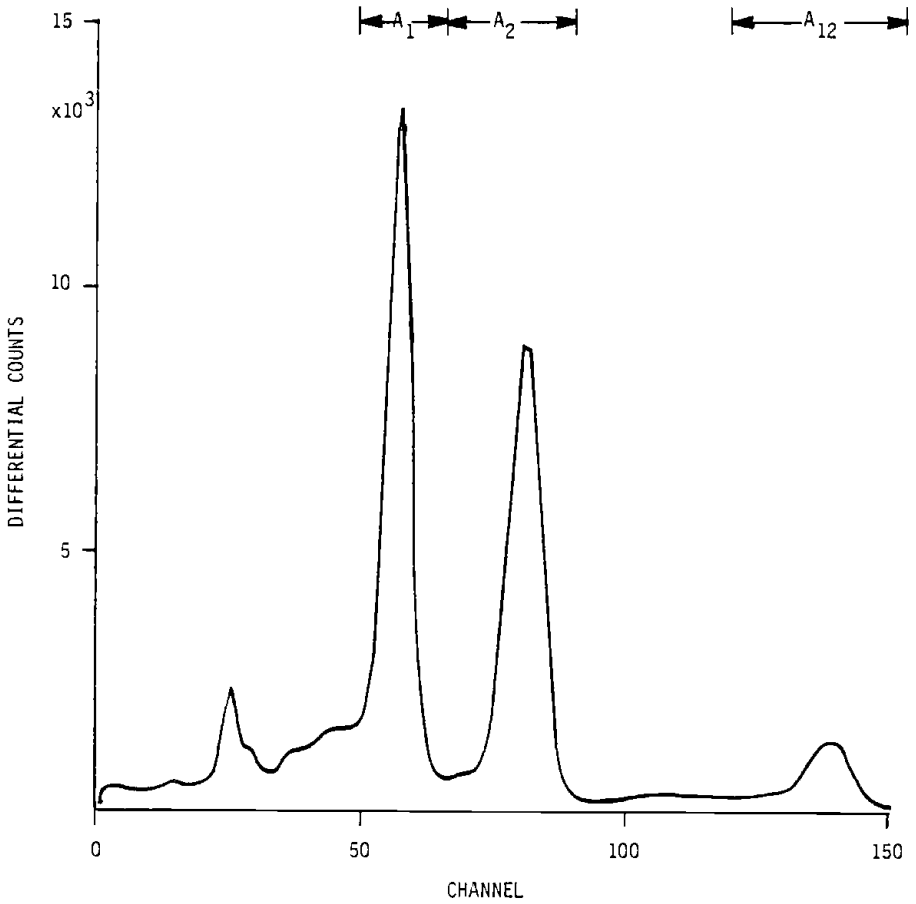


Figure 3. Pulse Height Spectrum of ^{111}In in Well-Thru NaI (Tl) Crystal Scintillation Detector with 0.3 cm-thick Brass Sleeve Around the ^{111}I Source and all Pb Shielding Removed.

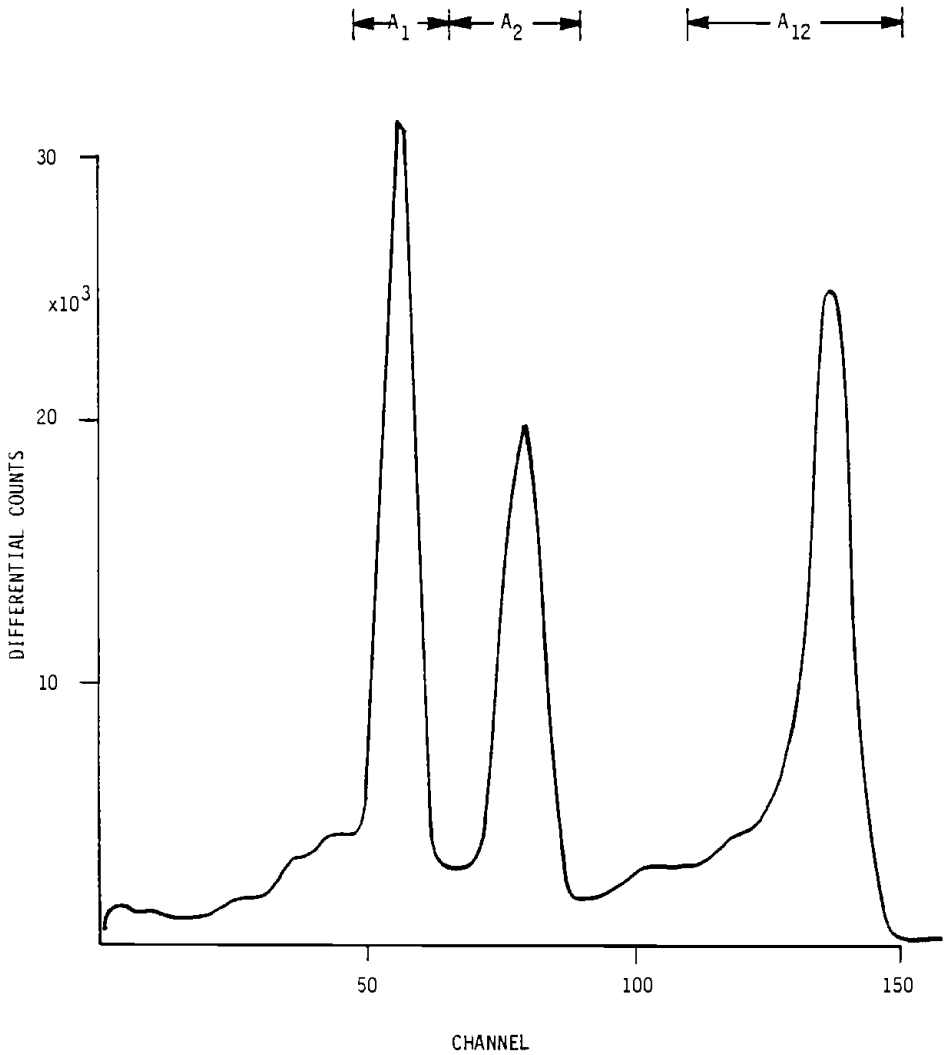


Figure 4. Pulse Height Spectrum of ^{111}In Outside of NaI (Tl) Crystal Scintillation Detector with 0.3 cm-thick Brass Sleeve around the ^{111}In Source.

Table 2. Specific Activity of ^{111}In Sources from Different Dilutions (Weights)

DATE MEASURED	WEIGHT	dpm/gm CORRECTED TO PST 0900 11-26-80	DIFFERENCE FROM NBS VALUE dpm/gm	%
11-26-80	1.035×10^{-3}	6.377×10^8	$- 0.001 \times 10^8$	0.00
12-3-80	1.035×10^{-3}	6.421×10^8	$+ 0.043 \times 10^8$	+ 0.67
12-8-80	2.031×10^{-3}	6.527×10^8	$+ 0.149 \times 10^8$	+ 2.34
12-8-80	3.997×10^{-3}	6.496×10^8	$+ 0.118 \times 10^8$	+ 1.85
12-8-80	7.740×10^{-3}	6.455×10^8	$+ 0.077 \times 10^8$	+ 1.21

and can be performed on a well-type NaI(Tl) system even by successive counts in a single-channel counting system; successively setting limits to measure A_1 , A_2 , A_{12} and T. For very accurate calibration, it is necessary to make corrections for the contribution of Pb X-rays produced by the photoelectric effect in the Pb shielding around the NaI(Tl) detector.

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