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ENVIRONMENTAL APPLICATIONS FOR AN INTRINSIC GERMANIUM WELL DETECTOR*

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ENVIRONMENTAL APPLICATIONS FOR AN INTRINSIC GERMANIUM WELL DETECTOR

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ABSTRACT

The overall performance of an intrinsic germanium well detector for ^{125}I measurements was investigated in a program of environmental surveillance. Concentrations of ^{125}I and ^{131}I were determined in thyroids of road-killed deer showing the highest activities of ^{125}I in the animals from the near vicinity of Oak Ridge National Laboratory. This demonstrates the utility of road-killed deer as a bioindicator for radioiodine around nuclear facilities.

INTRODUCTION

Intrinsic Ge or Ge(Li) well type detectors greatly increase the efficiency of high resolution gamma spectroscopy measurements in the determination of many of the important radioactive contaminants in the environment. This is especially important in the case of radionuclides that emit low energy gamma-rays or x-rays.

In this study, the analysis of ^{125}I in deer thyroids was performed using the well geometry of an intrinsic germanium detector attached to a computerized multichannel analyzer.

The overall performance of this detector for ^{125}I measurements was investigated and the results for ^{125}I and ^{131}I concentrations in deer thyroids were determined.

EXPERIMENTAL

Thyroid glands were extracted from white-tail deer killed on roads within the Oak Ridge reservation from May 19 to October 21, 1983. After extraction of the gland, fat and supporting tissue were removed and the glands placed into standard Petri dishes for ^{131}I measurements. After counting, the thyroids were lyophilized and the dry tissues were placed into plastic counting vials (13 mm internal diameter) for ^{125}I measurements.

The samples and ^{125}I standard solution were counted in identical counting vials in the well (16 mm dia x 45 mm length) of an intrinsic type germanium detector (23% relative efficiency at 25 cm as compared to that of a 3 x 3 - in. NaI (Tl) detector at 1.332 MeV). The spectrometer was operated at a gain of ∿ 0.1 keV/channel yielding an energy range of 20 to 400 keV.

2 All thyroid specimens containing 0.2 to 2.0 g of freeze-dried tissue in counting vials were counted for 25,000 to 150,000 sec., while the counting time for the ^{125}I standard solution was 1000 sec.

Concentrations of ^{125}I in thyroids were calculated from net integrated counts in 3 different regions of interest including: a) K_{α} x-ray peak of 27.2 + 27.5 keV, K_{β} x-ray peak of 31 keV and γ-ray peak of 33.5 keV, b) coincidence sum peaks at 54.8, 58.3, 62.8 and 66.3 keV, and c) of regions a) and b) together.

The absolute activity of ^{125}I standard solution was calculated by Eldridge's calculation (ref.1). The activity concentration of the original ^{125}I solution obtained from the New England Nuclear Co. was 5400 disintegrations per second/mL in approximately 0.02 molar NaOH solution. The test samples were prepared by dilution, so that the activity of the final working standard was 540 dps in 0.02 M NaOH, on September 28, 1983.

RESULTS AND DISCUSSION

The decay scheme of ^{125}I is unique because the nuclide decays 100% by electron capture followed by a 35.5 keV gamma ray transition. The K_{α} x-rays of 27.2 + 27.5 kev and K_{β} x-rays of 31 keV emitted after electron capture events in the ^{125}I are in coincidence with the photons and x-rays following internal conversion events emitted in the decay of the 35.5 keV excited state of ^{125}Te (ref.1). The coincident photons give rise to the characteristic gamma-ray spectrum which is partly shown in Figure 1.

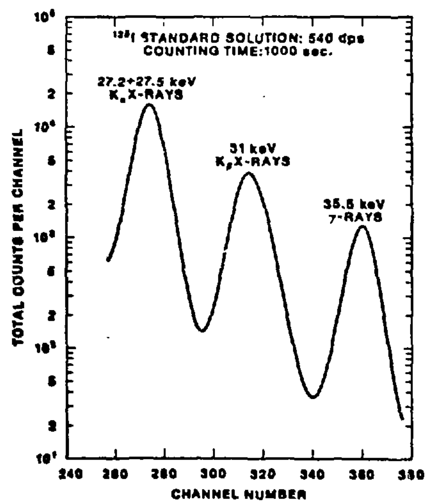

Fig. 1. Gamma-ray spectrum of 540 dps ^{125}I standard solution in the Ge well detector.

Table 1 shows the relative activity of a ^{125}I standard solution as a function of position in the well of the Ge detector. A standard solution containing 0.5 mL of ^{125}I was counted for 300 seconds at 8 different levels (0.5 cm intervals) from the bottom towards the top of the well. Fairly constant activity in the 27.4 keV K_{α} x-ray peak was observed for 0.5 to 2.5 cm of distance from the bottom of the well; the activity started to decrease at 3.0 cm and at 4.0 cm was less than 40% of the activity in lower positions of the counting vial in the well.

In Table 2 the relative activity of a ^{125}I standard solution as a function of dilution is presented. The results clearly show relatively constant activity (249 ± 5.7 cps) for 0.1 to 3.0 mL of ^{125}I standard solutions which gives a useful volume of almost 4 mL for samples in the counting vial. This means that up to 3 g of freeze-dried sample can be easily placed into the counting vial and counted without losing counting efficiency due to the sample geometry in the well.

TABLE 1
Relative activity of ^{125}I standard solution as a function of position in the well of a Ge detector.

Distance from the bottom of the well (cm)	cps
0.5	256
1.0	261
1.5	254
2.0	253
2.5	243
3.0	220
3.5	159
4.0	100

TABLE 2
Relative activity of ^{125}I standard solution in the well as a function of dilution.

Volume (mL)	cps
0.1	257
0.5	256
1.0	242
1.5	250
2.0	248
2.5	247
3.0	241
3.5	228

In Table 3 the ^{125}I and ^{131}I contents of the 29 deer thyroids of this study are shown. The highest quantities of ^{125}I were found in those animals (Nos. 65, 77, 80, 96) from the near vicinity of Oak Ridge National Laboratory (ORNL). This indicates that ORNL was the origin of ^{125}I in the environment.

High amounts of ^{131}I were also determined in thyroids of some deer collected within the Oak Ridge reservation after August 22, when ^{131}I was released from ORNL to the environment.

TABLE 3
^{125}I and ^{131}I contents in thyroids of 1983 road killed deer in Oak Ridge, TN.

Deer (No. sex)	Collection date	Weight (kg)	Age (year)	^{125}I (pCi/g D.W.)	^{131}I (pCi/g D.W.)
59 m	May 19	40	1	20	
60 f	May 20	54	3	23	
61 m	May 23	75	2	10	
65 f	Jun 2	42	1	105	
67 f	Jun 22	51	3	5.1	
77 m	Jul 16	47	1.5	97	
79 f	Jul 20	38	1	15	
BL3 f	Jul 20	13	Fawn	33	
80 f	Jul 21	54	3	42	
82 m	Aug 1	62	1.5	9.5	
85 m	Aug 11	35	1.5	3.6	
93 m	Aug 19	55	1.5	4.4	
96 f	Aug 22	38	1	35	
98 f	Aug 27	50	4	20	64
99 m	Aug 29	15	9 wks.	12	
101 f	Aug 31	41	1.5	5.8	27
102 m	Sep 2	19	10 wks.	2.2	200
103 f	Sep 7	64	3	16	4.0
104 f	Sep 7	25	16 wks.	2.6	47
105 m	Sep 9	55	2		13
108 m	Sep 12	71	2	11	210
109 m	Sep 12	20	16 wks.		49
111 m	Sep 14	23	16 wks.		17
NR1 m	Sep 25	53	2		<8
116 f	Sep 27	55	2		46
118 f	Sep 30	67	5		54
127 m	Oct 10	51	2		5.0
128 f	Oct 11	46	3		9.6
136 f	Oct 21	30	20 wks.		11

m = male
f = female
D.W. = freeze-dried weight

High ^{131}I concentrations significantly interfered with ^{125}I determinations in the same thyroid. In particular, the K_α x-rays of 29.5 + 29.8 keV (4% abundance) and 0.9% K_β x-rays of 33.6 keV in the decay mode of ^{131}I contributed to the ^{125}I spectrum at this energy region (27.2 + 27.5 keV K_α and 31 keV K_β x-rays).

Our intrinsic germanium well-type detector was found to be an extremely suitable device for ^{125}I measurements. The limitation of the sample quantity was not a disadvantage in the case of deer thyroids for which the lyophilization was performed as a preparation technique prior to the final analysis. The lyophilization greatly reduced the original size of the fresh tissue (70 to 80% of water) and enabled the accommodation of the entire freeze-dried thyroid in the counting vial inside the well of the detector.

The high efficiency of this well type Ge detector at energies below 100-keV can have applications in the measurement of other important radionuclides in the environment such as ^{129}I, ^{241}Am, ^{210}Pb, etc. (ref.2,3).

Several previous low-level radioactivity measurement methods can also be improved by the use of Ge or Ge(Li) well type detectors (ref.4,5,6).

Another useful application is the measurement of higher energy emitters in larger volume samples placed in standard containers around ("Marinelli" beakers) or on the top (Petri dishes) of the detector (ref.3).

With respect to the environmental contamination by radionuclides, this study clearly demonstrates the utility of road-killed deer as a bioindicator for radioiodine around nuclear facilities.

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