NSEC *Nuclear Science and Engineering Corporation*

REPORT

SURVEY OF IODINE-129 CONCENTRATIONS IN THYROID TISSUES

- - -

Progress Report [redacted]

Contract No. AT(30-1)-3049

Facsimile Price $ 2.60

Microfilm Price $ 1.07

Available from the
Office of Technical Services
Department of Commerce
Washington 25, D. C.

NSEC No.:	93
To:	U. S. Atomic Energy Commission Division of Biology and Medicine Washington 25, D. C.
Date:	May 17, 1963

SURVEY OF IODINE-129 CONCENTRATIONS IN THYROID TISSUES

Progress Report

Contract No. AT(30-1)-3049

To

United States Atomic Energy Commission

May 17, 1963

B. Keisch
R. C. Koch
A. Edelmann
R. R. Edwards
Niel Wald, M. D., Consultant

Nuclear Science and Engineering Corporation

TABLE OF CONTENTS

Progress Report - May 17, 1963

Survey of Iodine-129 Concentrations in Thyroid Tissues

I. INTRODUCTION

In accordance with the plan of work described in NSEC proposal 51-31-2004E, studies are under way of the iodine-129 concentrations in human and other mammalian thyroid tissues. Emphasis to date has been on (a) assembling a suite of thyroid specimens from a variety of sources, collected at times ranging from 1930 to the present; (b) development of I-129 analytical techniques to their ultimate sensitivity; and (c) analysis of a number of thyroid specimens of recent collection, in which I-129 concentrations were expected (and found) to be sufficiently high that ultimate sensitivity was not required in their determination. The limited supply of samples of less recent vintage, expected to contain less I-129, has been reserved pending complete determination of the optimum conditions (of neutron flux, sample size, etc.) for ultimate sensitivity measurements.

Success in applying the activation-analysis technique to natural iodine specimens has paralleled expectations (1) at thermal neutron flux values of the order of 10^{13}. Absolute sensitivities, in terms of I-129/I-127 ratios, may be limited by either fast neutron-induced reactions or third-order thermal neutron capture in I-127.

Background levels in the measurement of I-130 have been reduced by a factor of at least 10^2 as compared with those reported by the Argonne group (2) by use of a β-γ coincidence counting arrangement. This improvement in sensitivity has made it possible to obtain finite values for I-129 in analyses of individual adult human thyroid glands of recent collection date. Evaluation of the extent to which this may be the case with the glands of older collection date, or to which sensitivity limitations may be overcome by pooling of specimens, awaits results of the studies of the nuclear parameters of I-129/I-127 mixtures at higher neutron-flux values.

Progress in the various concurrent phases of the program is discussed in this report, and recommendations for future work are presented.

II. COLLECTION PROGRAM

Attempts have been made to obtain an array of human and animal thyroid specimens spanning the time period from the onset of the nuclear age to the present, in order that analyses will both reveal the pre-nuclear I-129 levels and yield some data on the rate at which this fission product is accumulating in the biosphere.

A. Human Specimens Collected 1962-63

Routine collections of surgical and post-mortem human thyroid specimens have been carried out for the past six months at five Pittsburgh hospitals and, with the cooperation of Dr. Merril Eisenbud, at the NYU-Bellevue Medical Center. These collections have yielded several hundred glands from pre-natal to geriatric, which are available for statistical studies of the current biospheric addition rates and/or of age variations in the thyroid I-129/I-127 ratio.

The collection of Dr. Henry N. Wellman of the Division of Radiological Health, U. S. Public Health Service, Cincinnati, has also been placed at our disposal. These specimens come from a widespread collection network, and have already been measured for I-131 shortly after collection.

B. Older Human Specimens

Preserved human thyroid sections, about seventy in number, have been obtained from a collection of Dr. Shields Warren of the New England Deaconess Hospital. These specimens were collected during the period 1945-50. Additional sources of specimens collected during 1945 or earlier are being sought.

C. Other Mammalian Specimens

Additional analytical samples of mammalian thyroid tissue covering a collection period extending from 1930 to 1962 have been assembled. These are listed below, with their sources and dates of collection:

1. Sheep thyroid glands, whole, frozen
   Source: Pittsburgh slaughterhouse
   Date: 1962
   Quantity: Approximately 50

2. Hog thyroid, desiccated
   Sources: Various pharmaceutical and meat packing companies
   Dates: 1957-1962
   Quantity: Several pounds

3. Thyroid nucleoprotein tablets (sheep)
   Source: Schieffelin and Co.
   Dates: 1930-1956
   Quantity: 1500 (iodine content 0.002 to 0.01 mg. each)

4. Thyroid tablets, 1/4 grain USP (hog)
   Source: Wilson Laboratories, Chicago
   Date: Pre-1945
   Quantity: 4000 tablets

III. EXPERIMENTAL PROCEDURE

Although I-129 is a radioactive nuclide, its extremely long half-life (1.6×10^7 years) results in a specific radioactivity of only 0.16 μc/mg. Furthermore, the low energy of its radiations (150-Kev β and 38-Kev γ) also make detection by ordinary counting techniques very insensitive. A reasonably large cross section (~27 barns) for thermal neutron capture, leading to the formation of the activation product, 12.6-hr I-130, makes the detection of I-129 by neutron-activation analysis highly practical (1).

A. Counting Procedures and Instrumentation

The activation of material containing I-129 for subsequent measurement of the I-130 produced leads to several interferences which cannot be circumvented except through the proper choice of the measurement method. For example, the normal presence of I-127 in the matrix leads to the formation of 25-minute I-128 by the (n, γ) reaction and to the formation of 13-day I-126 by the (n, 2n) reaction. The interference of the 25-minute I-128 may be reduced or eliminated through radioactive decay prior to measurement of the I-130. However, the 13.2-day I-126 which may be present must be discriminated against instrumentally. An examination of the decay schemes of I-126, I-128, and I-130 reveals that the maximum total energy of the gamma cascades in the decay of the respective nuclides is 1.41 Mev, 0.99 Mev, and 2.34 Mev. Therefore, it was anticipated that the use of a large diameter, well-type NaI gamma-scintillation detector, with a proper choice of gamma-energy discrimination, would permit the measurement of I-130 without serious interference from the other two iodine isotopes. The use of this type of detector reduces the Compton continuum and enhances the higher energy portion of a gamma spectrum like that of I-130; i.e., the larger the crystal, the greater the probability of total absorption of the energy of gamma cascades. The use of a 5-in diameter by 5-in long well-crystal yields ~40 percent detection efficiency for the radiations from the I-130 when the lower discrimination level is set at 1.6 Mev (a choice which excludes all but a very small amount of the radiation from the I-126). An upper level of discrimination (2.5 Mev) is also useful in reducing to a mini-

mum the counting rate due to cosmic radiation, which is rather high in a crystal of this size. However, without prohibitively excessive shielding, high background count rates (hundreds per minute) are still encountered.

To reduce the background to a truly low value and thus increase the sensitivity of the measurement technique, a β-γ coincidence counting system has been applied. This system (Figure 1, 2) utilizes the NaI crystal described above, which has a 1-in diameter well, 3-in deep. A thin β-sensitive plastic phosphor, with a configuration such that a sample is nearly completely surrounded by a 1/16-in thick layer of the phosphor, is positioned within the well. A lucite light pipe and a small photomultiplier tube complete the detector assembly. Coincidence conditions are imposed on the output of the β-phosphor and the discriminated output of the large gamma scintillator by a coincidence circuit with a resolving time of approximately 0.08 microseconds, and the coincidence count rate is measured. The overall detection efficiency for I-130 counted in this system varies from 10 percent to 40 percent, depending on the sample thickness and the choice of the lower gamma-energy discrimination level. Typical samples usually exhibit efficiencies in the lower portion of this range. The background counting rate for the system is of the order of only 0.06 - 0.07 counts per minute. Thus, a relatively small sacrifice in efficiency, from 40 percent to ~15 percent, results in an extremely valuable reduction in background counting rate of 3 orders of magnitude, and the sensitivity of the measurement is enhanced by a factor of 100 or more.

The ultimate limit of detection of I-129 in a given sample depends on a number of complex factors. With the experimental procedures and counting arrangement cited, and with neutron irradiations in a flux of $\sim 10^{13}$ neutrons/cm^2 sec, $\sim 10^{-12}$ grams of I-129 may be <u>easily</u> measured in a typical sample with an ultimate sensitivity approaching 10^{-13} grams. In principle, an increase in thermal neutron flux will improve the sensitivity proportionately. However, a number of additional factors must be considered.

1. <u>Production of I-126.</u> As has been already noted, the presence of a finite quantity of stable I-127 in a sample leads to the production of

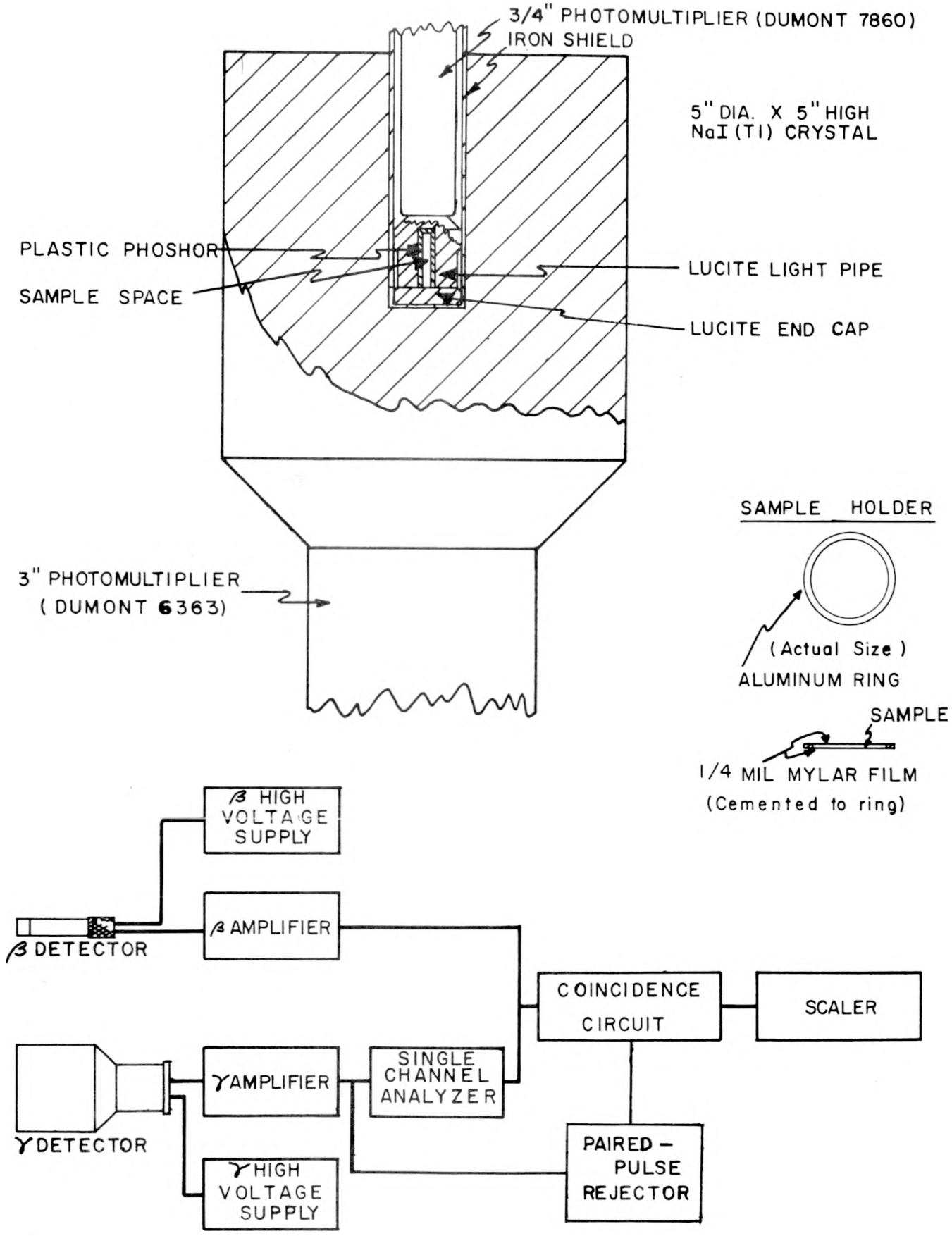
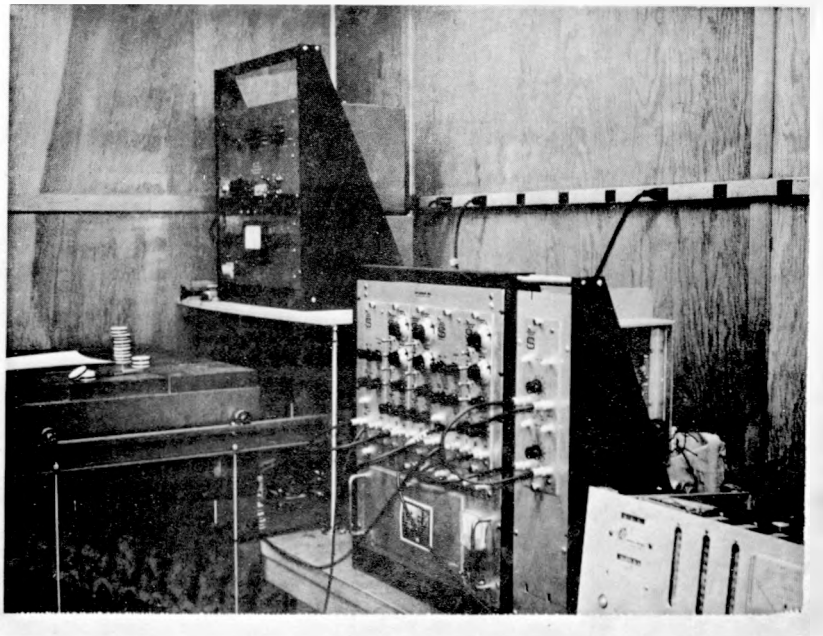


FIGURE 1. β–TOTAL γ COINCIDENCE COUNTING ARRANGEMENT



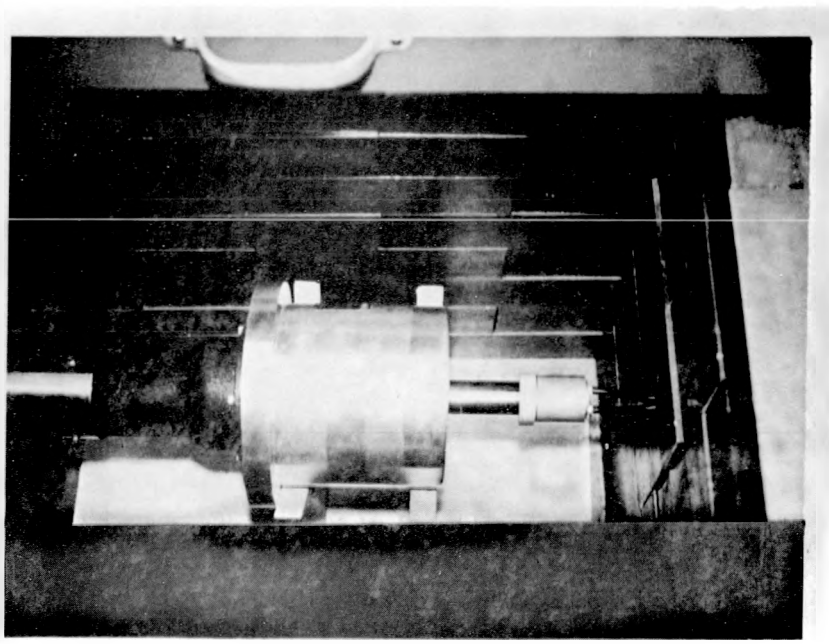


FIGURE 2 β - TOTAL γ COINCIDENCE COUNTING EQUIPMENT

I-126 via the (n, 2n) reaction. This reaction occurs to an appreciable extent whenever a significant fraction of the neutrons have an energy greater than 9 Mev. Depending on the discrimination level at which the gamma counting circuit is operated, a certain I-126 background count rate is observed for each sample. It is this count rate which places a limit on the sensitivity of the detection system for counting I-130.

Even with complete discrimination against the gamma rays of energies known to be associated with the decay of I-126, such a count rate is observed. This is true particularly for samples containing large quantities of I-126. The cause of this phenomenon is related to the time resolution of the electronic circuits since, as is common with high counting rates, two, accidentally coincidental, gamma rays are occasionally observed as a single, higher-energy event. Additional electronics designed to minimize this effect have been added to the basic circuitry. The additional circuitry is designed to reject such "paired pulses" by examination of the duration of the single pulse observed. A "paired pulse" is longer in duration than a normal pulse. Use of this technique results in a three- or fourfold reduction in the I-126 background count rate.

As a further method if improving the situation, arrangements have been made for use of the MIT reactor, which is reported to provide a higher ratio of thermal to fast flux. It is estimated that when the improved facility is used, the sensitivity will be increased to a point where the theoretical lower limit for detection of I-129 will be approached. This limit is controlled by the following.

2. Multiple Neutron Capture in I-127. Iodine-130 can be produced in an as yet undetermined amount through the successive capture of three neutrons by the stable I-127 present in a sample. The neutron-activation cross section of I-128 is unknown and could be as high as 1000 barns. The radioactivity of I-130 produced by triple neutron capture after the first three hours of irradiation of one gram of I-127 will follow the approximate equation (see Appendix A):

$$A_{130} = 1.9\,(10^{-21})\,\phi^{3}\,\sigma_{128}\,t(1-e^{-\lambda_{130}t}), \qquad (1)$$

where A_{130} is the disintegration rate of the I-130 produced from I-127 at the time of removal from the neutron flux; ϕ is the neutron flux; σ_{128} is the neutron-activation cross section of I-128; t is the time of irradiation; and λ_{130} is the decay constant of I-130. The disintegration rate of I-130 produced from the I-129 originally in the sample follows the equation:

$$A_{130} = 2.7\,(10^{-23})\,N_{129}\,\phi(1-e^{-\lambda_{130}t}), \qquad (2)$$

where N_{129} is the number of atoms of I-129 present, and the other symbols have the same significance as in Equation (1). The interference levels for various values of σ_{128} have been calculated assuming a 12-hour irradiation period. Table I shows the calculated approximate equivalent values of the I-129 to I-127 ratio simulated by triple neutron capture in I-127.

TABLE I

CALCULATED INTERFERENCE LEVELS DUE TO TRIPLE NEUTRON CAPTURE IN I-127
(12-hour irradiation)

σ_{128} (barns)	I-129/I-127	
	$\phi = 10^{13}$	$\phi = 10^{14}$
10^3	6×10^{-11}	6×10^{-9}
10^2	6×10^{-12}	6×10^{-10}
10	6×10^{-13}	6×10^{-11}

For a typical sample of iodine extracted from a thyroid gland, $\sim10^{-2}$ grams of I-127 is irradiated. Detection of $\sim10^{-13}$ grams of I-129 (maximum sensitivity for $\phi = 10^{13}$) would give a ratio of 10^{-11} g I-129/g I-127. Inspection of Table I reveals that, for a flux of 10^{13} neutrons/cm^2 sec, only a relatively high value for σ_{128} will produce a detectable amount of I-130 under these conditions. However, for a flux of 10^{14}, and with an expected ultimate sensitivity approaching 10^{-14} grams of I-129, a value of 10^{-12} for the ratio I-129/I-127 would be obtained and even a relatively low value of σ_{128} will

cause interference. It is, therefore, imperative that the value of σ_{128} be determined in order to ascertain whether multiple neutron capture will alter the lower limit of the sensitivity of the method for I-129 detection and, if so, to what degree.

An experiment has been performed to determine the value for the cross section of I-128. A series of samples of iodine were prepared by careful purification of an iodine-containing compound which is known to predate the atomic era. These samples were irradiated with flux monitors for varying periods of time at a flux of $\sim 2(10^{14})$ neutron/cm^2 sec. Determinations of the I-130 produced in each of these samples enabled the calculation of σ_{128} by means of the exact equation given in Appendix A. The tentative value obtained indicates that σ_{128} is of the order of 100 barns. An additional experiment has been scheduled for the immediate future in order to confirm the result obtained in the first experiment.

Assuming that the value of σ_{128} is indeed ~100 barns, then, as Table I reveals, the ultimate sensitivity for the ratio measurement (I-129/I-127) is approximately 6×10^{-12} with the currently employed methods.

B. Chemical Procedures

1. Pre-irradiation Chemistry

a. Extraction of Iodine from Thyroid Glands and Thyroid Powder. The procedure used for extraction of iodine in an inorganic form from the organic material of a whole thyroid gland or from desiccated thyroid powder involves the digestion of the organic material with sodium hydroxide. Typically, the procedure used is as follows.

A whole human thyroid gland (or approximately 10-20 grams of dry thyroid powder) is placed in a 5-in porcelain evaporating dish, approximately 2-3 grams of sodium bisulfite and 30 ml of 6 N NaOH solution are added, and the mixture is slowly (~2 hours) evaporated to dryness. The material is transferred to a 100-ml nickel crucible which is then heated gently with a Meker burner until combustion of the organic material is complete. The crucible is then placed in a muffle furnace at ~1000° C for 2-3 minutes. Upon removal from the furnace, the crucible is allowed to cool

until no red glow is visible and is then quenched in sufficient deionized water to cover it. The solidified melt is then loosened from the crucible, and the latter is removed from the solution and rinsed into it. The dissolution of the melt is completed by heating the solution on a hot plate. The solution is carefully acidified with nitric acid, and 20-30 ml of 10 percent sulfurous acid is added. Any undissolved solids remaining are removed by filtering the solution through a millipore filter.

A check on a typical yield of the above procedure was provided in the following way. A weighed sample of thyroid powder was digested in the manner described above and the iodine precipitated as silver iodide. The silver iodide after careful washing and drying, was weighed, and the percent of iodine in the original sample was calculated. This result, 0.30 percent, agreed very well with that obtained on a duplicate sample by a recognized microanalytical laboratory using standard techniques for organic iodine analysis. The independent result, 0.31 percent, indicates that the iodine extraction method used here is very nearly quantitative.

The total iodine extracted from whole human thyroid glands has been, on the average, approximately that expected for normal glands.

b. Purification of Iodine and Preparation for Irradiation. Extremely careful purification of the iodine with respect to two contaminants, cesium and uranium, is necessary prior to irradiation. The presence of either of these can lead to the formation of I-130 in a neutron flux. An (n, α) reaction on cesium or the fission of uranium can produce I-130 in sufficient quantities to vitiate the results of the activation, particularly when there is only a very small quantity of I-129 present.

The solution obtained in section (a) above is purified by two cycles of extraction as elemental iodine and back-extraction as iodide as follows.

Sodium nitrite solution is added to the solution resulting from the dissolution of the organic material until the brown color of I_2 is observed. The elemental iodine is then extracted into carbon tetrachloride.

After the organic phase is washed with deionized water, the iodine is back-extracted into a dilute solution of sulfurous acid, oxidized with sodium nitrite again, and the cycle is repeated. The final aqueous solution is boiled to remove carbon tetrachloride, allowed to cool, and palladous chloride ($PdCl_2$) is added to precipitate palladium iodide. The precipitate is removed by filtering through a fritted glass funnel, and is then washed with several portions of deionized water followed by alcohol and ether. The frit and precipitate are dried by placing for 15 minutes or more in an evacuated vacuum desiccator.

The dried palladium iodide is subsequently placed in one leg of a clean quartz* U-tube which is then evacuated and sealed. The compound is decomposed by gentle heating with a torch and the iodine distills into the other leg of the tube which is cooled in ice-water. The second leg, now containing elemental iodine is then sealed off and is ready for irradiation.

In the event that a thyroid gland contains an amount of iodine which is too small for handling in the above procedure (i.e., <1 mg as in the case of some pediatric samples), the following procedure is used for the preparation of the sample for irradiation. Five milligrams of silver, as a nitrate solution, is added to the final aqueous phase and sufficient dilute HCl is added to precipitate all of the silver. The silver chloride-iodide precipitate is filtered, washed, and dried in an oven at 110° C. The dry solid is placed in one arm of a clean quartz* U-tube with ~50 mg of aluminum wire, and the U-tube is evacuated and sealed. The solid mixture is heated gently with a torch until a substantial amount of aluminum chloride is distilled into the second leg of the U-tube which is cooled in an ice bath. The leg containing iodine and aluminum chloride is sealed off and is ready for irradiation.

2. Post-irradiation Chemistry and Preparation for Counting.

There are several interfering radioactivities which may be produced from impurities in the iodine or in the quartz irradiation capsule and which would be detected in the coincidence system described above. The most important

---

*The quartz tubes are cleaned by soaking in a solution consisting of ~20% HNO_3 and ~5% HF for 10-15 minutes and rinsing thoroughly with deionized water.

of these impurities is 36-hour Br-82, because it is most likely to survive the pre-irradiation purification chemistry by virtue of its chemical similarity to iodine. The method described below is specifically designed to decontaminate the sample from any Br-82 which may be present, but in so doing will also remove to an even greater extent, any other impurities, such as Na-24 or K-42, which may be present.

Following neutron-irradiation, the quartz capsules are broken open after thorough cleaning of the outside and immediately submerged in a solution of sodium bisulfite. For samples containing only a small amount of iodine, there is a danger of loss due to volatilization. These samples are opened in the apparatus shown in Figure 3. The vertical section of the tube is first filled with sodium bisulfite. The quartz tube is scratched and placed with the scratch mark over the joint. The joint is closed and held closed with a clamp (not shown). The joint is then flexed to break the capsule and the apparatus manipulated (without opening) so that the quartz is thoroughly rinsed with the bisulfite solution.

Following dissolution of the iodine, ~5-10 mg of bromine is added as a hold-back carrier in the form of a solution of sodium bromide, the iodide is oxidized to elemental iodine with nitrite, and the I_2 is extracted into carbon tetrachloride. A cyclic procedure similar to that described for the pre-irradiation chemistry is followed, except that bromine hold-back carrier is added to each aqueous extract (except the last) and three complete cycles are used*. Finally, the iodine is precipitated as silver iodide which is removed by filtering, washed, and dried. The silver iodide is weighed on a mylar film sample holder (Figure 1), evenly spread, and covered with another mylar film. The decay of the I-130 is followed for several days and a slight "tail" due to I-126 is usually obtained and subtracted. The area of the sample is measured and a self-absorption correction applied. The I-127 (stable) content of the sample is calculated from the weight of the silver iodide.

*The identical procedure was used on a sample which contained a deliberately added, known amount of bromine in order to examine the effectiveness of the decontamination procedure. No Br-82 was detectable in the purified sample and a decontamination factor of $2(10^5)$ was conservatively estimated as a lower limit.

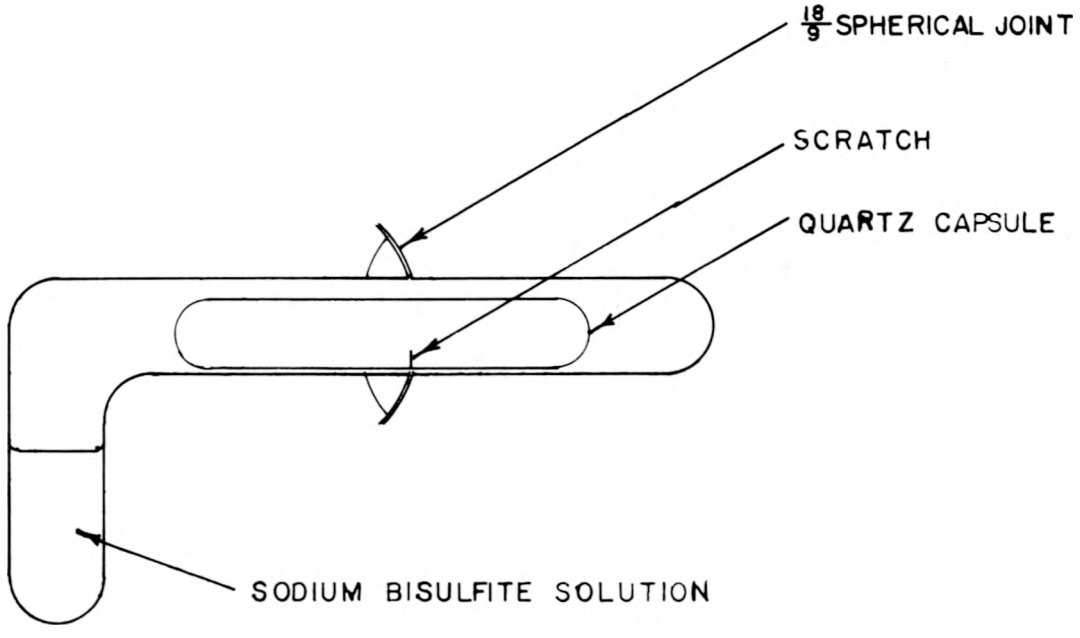


FIGURE 3 CAPSULE-OPENING APPARATUS

For samples containing an amount of iodine too small for precipitation or accurate weighing, a small amount of iodide carrier is added prior to the first extraction. The I-127 (stable) content of the sample is then obtained by comparison of its 13-day I-126 activity with other, larger, samples in the same irradiation after the I-130 has completely decayed.

Two comparators and two blank samples are run with each irradiation. The blanks are processed, after the addition of iodine carrier, in the same manner as the samples. These normally show no I-130 radioactivity.

The comparators consist of a known amount of I-129 in the form of NaI from a stock solution which has been irradiated with no prior treatment. After irradiation, a known amount of iodine carrier is added, and one purification cycle is performed to remove the Na-24 which is present. Silver iodide is precipitated and weighed, and the yield and self-absorption corrections are calculated in the usual manner.

IV. RESULTS

Thirty-four individual whole human thyroid glands and four samples of desiccated hog thyroid have been successfully analyzed thus far in this program. All of the whole human thyroid samples were current, all but two having been obtained from autopsies at three hospitals in the Pittsburgh area during late 1962 and early 1963. (The two exceptions are samples obtained from the NYU - Bellevue Medical Center.) The thyroid powders were of recent (1957-1962) manufacture from hog thyroid by Parke-Davis and Company, Detroit, Michigan, by the Pharmaceutical Division of Wilson Laboratories, Chicago, Illinois, and by Canada Packers Limited of Toronto, Canada.

The results of the analyses of the human thyroids are shown in Table II. The isotope ratio I-129/I-127 varies from 8×10^{-11} to 1.5×10^{-8} for these human thyroid samples, with an average of 2.7×10^{-9}. The values for the hog thyroid tissues shown in Table III range from 4.8×10^{-10} to 3.1×10^{-9} for this ratio. The ages of the subjects from whom the thyroid glands were taken and the dates of manufacture of the thyroid powder samples are given. There were a few adult thyroid samples that exhibited values orders of magnitude higher than those shown in the table (up to $\sim 10^{-6}$ g I-129/g I-127). Judgment on the validity of these values is being withheld until such time as Dr. Wald may examine the case histories of these subjects.

The data show clearly that current biospheric I-129 levels are higher by factors of at least 10^4 to 10^6 than those anticipated (1) as a result of spontaneous fission and cosmic-ray production processes. (A sample of verified pre-atomic era potassium iodide was analyzed and no detectable I-129 was found. The estimated upper limit was 9×10^{-11} g I-129/g I-127.) The limited range of collection dates of the human specimens analyzed to date prohibits conclusions as to the current rates of biospheric accumulation. Similarly, the small number of samples so far analyzed is not considered to be statistically adequate for conclusions regarding variation of I-129 level with age of the individual. However, the averages and medians of age groups by

decade are shown graphically in Figure 4. If the variation in isotope ratio with age is borne out in the analyses of a large number of additional samples, a considerable dietary or other effect is certainly suggested.

Geographical variations, and perhaps a biospheric increase in the period 1957-1962, are suggested by the results on the desiccated hog thyroid samples. Since these are normally prepared from large numbers of glands collected in a short period of time at a given location, individual differences should be pretty well eliminated. Again, however, it is clear that additional analyses will be required to justify any firm conclusions.

A calculation, assuming a total iodine content of 15 mg in the adult thyroid, shows the total I-129 content of the human thyroid specimens ranging from 1.2 to 220 picograms (2×10^{-4} to 3×10^{-2} picocuries) per adult gland, clearly demonstrating the sensitivity of the analytical method at its present level of refinement.

TABLE II

CURRENT I-129 CONTENT OF HUMAN THYROID TISSUES

Sample	Date of Collection	Age of Subject	Isotope Ratio I-129/I-127 ($\times 10^9$)
1	9/62	48	6.5 ± 0.7
2	9/62	43	2.6 ± 0.2
3	10/62	42	0.2 ± 0.1
4	9/62	74	0.09 ± 0.06
5	9/62	57	0.26 ± 0.09
6	12/62	50	4.3 ± 0.2
7	10/62	52	2.5 ± 0.4
8	9/62	44	1.1 ± 0.2
9	11/62	37	2.4 ± 0.2
10	12/62	38	3.0 ± 0.5
11	9/62	46	0.37 ± 0.07
12	10/62	34	3.2 ± 0.2
13	9/62	52	0.20 ± 0.03
14	10/62	60	0.54 ± 0.13
15	10/62	55	0.24 ± 0.10
16	12/62	58	8.1 ± 0.4
17	9/62	52	0.08 ± 0.01
18	9/62	81	2.8 ± 0.1
19	9/62	49	9.4 ± 0.6
20	9/62	61	1.2 ± 0.5
21	2/63	62	<1
22	3/63	48	0.8 ± 0.1
23	2/63	47	2.5 ± 0.5
24	2/63	69	1.0 ± 0.1
25	2/63	84	<0.5
26	2/63	62	2.5 ± 0.5
27	2/63	50	15 ± 3
28	3/63	48	1.9 ± 0.9
29	3/63	75	5 ± 3
30	9/62	85	<2
31	12/62	64	5 ± 3
32	4/63	10	5.4 ± 1.5
33	5/62	>25[(a)]	2.3 ± 0.4
34	5/62	>25[(a)]	1.6 ± 0.4

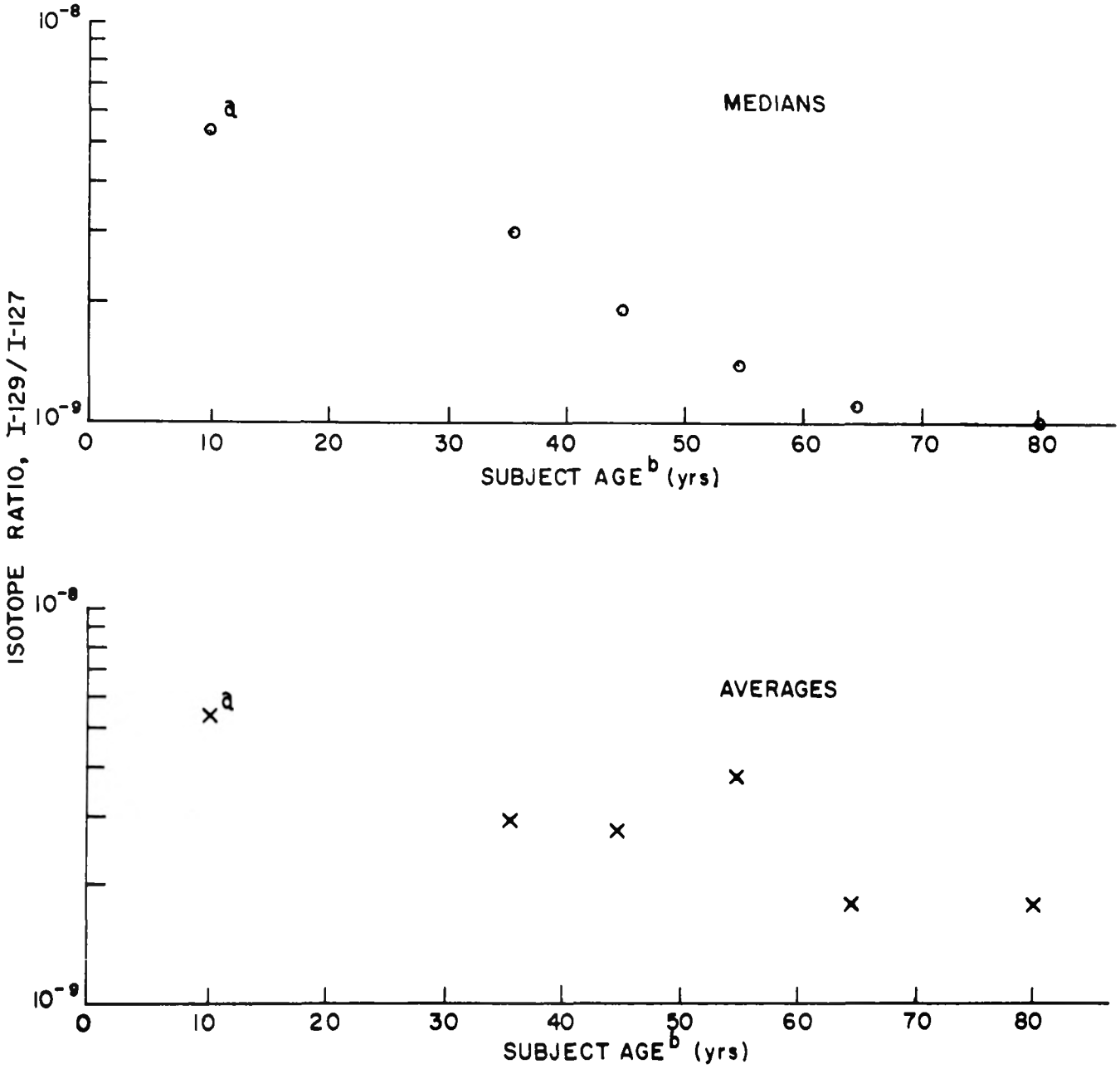
(a) NYU samples.

TABLE III

I-129 CONTENT OF HOG THYROID POWDERS

Sample	Date of Preparation	Isotope Ratio[(a)] I-129/I-127 ($\times 10^9$)
Parke Davis	1962	3.1 ± 0.6
Parke Davis	1957-58	0.48 ± 0.05
Wilson	1962	1.1 ± 0.1
Canadian Packers Ltd.	1962	0.61 ± 0.06

(a) Averages of duplicate determinations. Maximum spread of duplicates ~ 25 percent.



a - One value only.

b - Grouped in ranges <39, 40-49, 50-59, 60-69, >70 and plotted as average age of the extreme groups or mid-point of the other groups.

FIGURE 4
GRAPHIC PRESENTATION OF I-129 CONTENT OF CURRENT THYROIDS

V. CONTRIBUTION OF IODINE-129 TO TOTAL THYROID RADIATION DOSE

In the absence of extraneous contamination, body tissues receive a relatively constant, irreducible radiation dose from K-40, C-14, and H-3, with additional variable contributions from the naturally-occurring radioactive series elements. Except for astatine, none of the natural radioelements are expected to concentrate in thyroid tissue, and the astatine isotopes are formed in quite low yield and are too short-lived (10^{-4} sec to 2 sec) to concentrate efficiently.

Assuming then that K-40, C-14, and H-3 are the principal sources of ionizing radiation to thyroid tissue, and assuming the tissue to be 80 percent water, Table IV shows the expected activity in normal adult thyroid tissue from these naturally-occurring radionuclides, as compared with observed values for I-129.

TABLE IV

ESTIMATED ACTIVITY LEVELS OF NATURALLY-OCCURRING RADIOELEMENTS IN THYROID TISSUE: COMPARISON WITH I-129 ACTIVITY

Element	g Element/gland	Specific Activity dpm/g element	Total Activity pc/gland
C	2.0	15 (C-14)	13
H	2.2	0.009 (H-3)	0.009
K	0.03	1780 (K-40)	24
I	0.015	1.4 (I-129)	~0.01

Without further analysis, it is clear from these rough estimates that I-129 at current biospheric levels is making a minor contribution to the total thyroid tissue dose. If as indicated in the limiting estimates presented in Appendix B, the localized doses due to I-129 are in the μrad per year range, C-14 (with a similar decay energy) delivers average dose rates ~ 10^4 times those of I-129, thus in the 10 millirad per year range. With its gamma radiation and higher energy β particles, the K-40 contribution to the thyroid tissue dose will be a fraction of the total decay energy dissipated, but still near the order of magnitude of that of C-14.

Appendix A

I-130 ACTIVITY PRODUCED BY TRIPLE NEUTRON CAPTURE IN I-127

Part I - Exact Solution

The solution of the equations (ref. p. 137 et seq., Friedlander and Kennedy, Nuclear and Radiochemistry, John Wiley and Sons, New York, 1955) for the transformation of a chain of nuclides in a neutron flux has been given as

$$N_n = C_1 e^{-\Lambda_1 t} + C_2 e^{-\Lambda_2 t} + \cdots\cdots C_n e^{-\Lambda_n t} \qquad (1)$$

where $$C_1 = \frac{\Lambda_1^* \Lambda_2^* \cdots\cdots \Lambda_{n-1}^*}{(\Lambda_2-\Lambda_1)(\Lambda_3-\Lambda_1)\cdots\cdots(\Lambda_n-\Lambda_1)} N_1^o \qquad (2)$$

$$C_2 = \frac{\Lambda_1^* \Lambda_2^* \cdots\cdots \Lambda_{n-1}^*}{(\Lambda_1-\Lambda_2)(\Lambda_3-\Lambda_2)\cdots\cdots(\Lambda_n-\Lambda_2)} N_1^o, \text{ etc.} \qquad (3)$$

and N_n = number of atoms of a radioactive species of decay constant λ_n (sec^{-1}),

$\Lambda_n = \lambda_n + \phi \sigma_n$ (ϕ = neutron flux (cm^{-2} sec^{-1}), σ = total neutron reaction cross section),

t = time of irradiation (sec), and

$\Lambda_n^* = \lambda_n^* + \phi \sigma_n^*$,

where the asterisks indicate that only that part of the decay constant leading to formation of the next chain member is to be considered.

In the case of the reactions

$$I^{127} \xrightarrow[n,\gamma]{\sigma=6.7b} I^{128} \xrightarrow[n,\gamma]{\sigma=\sigma_{128}} I^{129} \xrightarrow[n,\gamma]{\sigma=26b} I^{130}$$

$$I^{128} \xrightarrow{\beta^-,\ t_{\frac{1}{2}}=28\ \text{min}} \qquad I^{129} \xrightarrow{\beta^-,\ t_{\frac{1}{2}}=1.6\times10^{7}\ \text{yrs}} \qquad I^{130} \xrightarrow{\beta^-,\ t_{\frac{1}{2}}=12.5\ \text{hrs}}$$

we obtain for equation (1) after use of the general equation for radioactive decay ($A = N\lambda$), and with the use of subscripts indicating particular nuclides;

$$A_{130} = N_{130}\,\lambda_{130} = \lambda_{130}\,N^{o}_{127}\,\Lambda^{*}_{127}\,\Lambda^{*}_{128}\,\Lambda^{*}_{129}\left[\frac{e^{-\Lambda_{127}t}}{(\Lambda_{128}-\Lambda_{127})(\Lambda_{129}-\Lambda_{127})(\Lambda_{130}-\Lambda_{127})}+\right.$$

$$\frac{e^{-\Lambda_{128}t}}{(\Lambda_{127}-\Lambda_{128})(\Lambda_{129}-\Lambda_{128})(\Lambda_{130}-\Lambda_{128})}+\frac{e^{-\Lambda_{129}t}}{(\Lambda_{127}-\Lambda_{129})(\Lambda_{128}-\Lambda_{129})(\Lambda_{130}-\Lambda_{129})}+$$

$$\left.\frac{e^{-\Lambda_{130}t}}{(\Lambda_{127}-\Lambda_{130})(\Lambda_{128}-\Lambda_{130})(\Lambda_{129}-\Lambda_{130})}\right] \tag{4}$$

Rearranging and adding plus and minus unity to the first term one obtains:

$$A_{130} = \lambda_{130}\,N^{o}_{127}\,\Lambda^{*}_{127}\,\Lambda^{*}_{128}\,\Lambda^{*}_{129}\left[\frac{(1-e^{-\Lambda_{129}t})-(1-e^{-\Lambda_{127}t})}{(\Lambda_{128}-\Lambda_{127})(\Lambda_{129}-\Lambda_{127})(\Lambda_{130}-\Lambda_{127})}+\right.$$

$$\left.\frac{e^{-\Lambda_{128}t}}{(\Lambda_{127}-\Lambda_{129})(\Lambda_{128}-\Lambda_{129})(\Lambda_{130}-\Lambda_{129})}+\frac{e^{-\Lambda_{130}t}}{(\Lambda_{127}-\Lambda_{130})(\Lambda_{128}-\Lambda_{130})(\Lambda_{129}-\Lambda_{130})}\right] \tag{5}$$

The following substitutions may be made:

$$\sigma_{127} = 6.7\ (10^{-24})\ \text{cm}^2$$

$$\sigma_{128} = \sigma_{128}$$

$$\sigma_{129} = 2.7\ (10^{-23})\ \text{cm}^2$$

$$\Lambda^{*}_{127} = \Lambda_{127} = \phi\,\sigma_{127} \quad \text{(because } \lambda_{127} = 0\text{)}$$

$$\Lambda^{*}_{128} = \phi\,\sigma_{128}$$

$$\Lambda_{128} = \lambda_{128} \quad \text{(because } \lambda_{128} >> \phi\,\sigma_{128} \text{ for ordinary values of } \phi\text{)}$$

$$\Lambda^{*}_{129} = \Lambda_{129} = \phi\,\sigma_{129} \quad \text{(because } \lambda_{127} << \phi\,\sigma_{129} \text{ for ordinary values of } \phi\text{)}$$

$$\Lambda_{130} = \lambda_{130} \quad \text{(because } \lambda_{130} >> \phi\,\sigma_{130} \text{ for ordinary values of } \phi\text{)}$$

and for an irradiation time (t) of less than 4 months,

$$1-e^{-\Lambda_{129}t} = \Lambda_{129}t$$

$$1-e^{-\Lambda_{127}t} = \Lambda_{127}t\,.$$

Making these substitutions and noting that

$$\Lambda_{130} \text{ or } \Lambda_{128} >> \Lambda_{127} \text{ or } \Lambda_{129} \text{ one obtains}$$

$$A_{130} = \lambda_{130} N^{o}_{127} \phi^{3} \sigma_{127} \sigma_{128} \sigma_{129} \left[\frac{t\,(\Lambda_{129} - \Lambda_{127})}{(\lambda_{128})(\lambda_{130})(\Lambda_{129}-\Lambda_{127})} + \frac{e^{-\lambda_{128}t}}{\lambda^{2}_{128}\,(\lambda_{130}-\lambda_{128})} + \frac{e^{-\lambda_{130}t}}{\lambda^{2}_{130}\,(\lambda_{128}-\lambda_{130})} \right] \qquad (6) \text{ or}$$

$$A_{130} = \lambda_{130} N^{o}_{127} \phi^{3} \sigma_{127} \sigma_{128} \sigma_{129} \left[\frac{t}{\lambda_{128}\lambda_{130}} + \frac{e^{-\lambda_{128}t}}{\lambda^{2}_{128}\,(\lambda_{130}-\lambda_{128})} + \frac{e^{-\lambda_{130}t}}{\lambda^{2}_{130}(\lambda_{128}-\lambda_{130})} \right] \qquad (7)$$

Part II - Approximate Solution

To a first approximation, for a 12-hour irradiation, one may consider that the nuclide I_{128} ($t_{\frac{1}{2}}$ = 28 min.) is present at an equilibrium concentration (true after ~3 hours).

The number of atoms of I-128 at equilibrium may be expressed as

$$N_{128} = \frac{N^{o}_{127} \phi \sigma_{127}}{\lambda_{128}} . \qquad (8)$$

After 3 hours (but less than $\sim 10^{6}$ years, the number of atoms of I-129 produced will follow the equation

$$N_{129} = N_{128} \phi \sigma_{128} t = \frac{N^{o}_{127} \phi^{2} \sigma_{127} \sigma_{128} t}{\lambda_{128}} \qquad (9)$$

and the activity of the I-130 produced will equal

$$A_{130} = N_{129} \phi \sigma_{129} (1-e^{-\lambda_{130} t})$$

$$= \frac{N^{o}_{127} \phi^{3} \sigma_{127} \sigma_{128} \sigma_{129} t (1-e^{-\lambda_{130} t})}{\lambda_{128}} . \qquad (10)$$

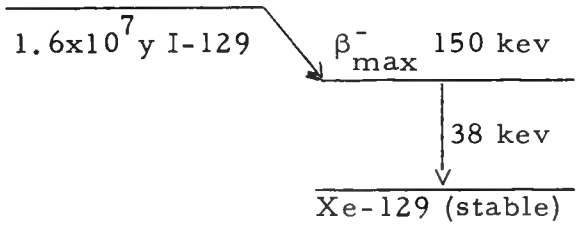
Substituting the known values of the constants in the above and for one gram of I-127 one obtains

$$A_{130} = 1.9 (10^{-21}) \phi^{3} \sigma_{128} t (1-e^{-\lambda_{130} t}) . \qquad (11)$$

Appendix B

UPPER-LIMIT DOSIMETRY OF IODINE-129

The decay scheme of I-129, as reported in the Nuclear Data Sheets (NRC 61-1-101), involves the emission of a 150 kev β^-_{max} particle, followed by a 38 kev quantum transition to the ground state of Xe-129:



As a highly-forbidden β transition is indicated by the small decay constant of I-129, the average β energy is probably not more than 20 percent of the maximum energy, so that $\overline{E}_\beta \cong 30$ kev as an upper limit.

The quantum transition in the Xe-129 daughter is reported to show a K-conversion coefficient $\alpha_K = 22$, and a K/L conversion ratio of about 10, indicating virtually complete internal conversion in the transition. The K-conversion events will yield ~5 kev electrons in ~95 percent of the transitions, along with Xe K-X rays at ~30 kev; the L-conversion events will yield ~30 kev electrons, along with Xe L-X rays in the remaining 5 percent or so of the events.

Neglecting completely the K- and L-fluorescence yield values, i.e., assuming that the total transition energy (except for neutrinos) appears as soft electrons, the energy absorbed in tissue adjacent to a disintegration event would be 68 kev per disintegration. The range of the electrons involved would be ~0.01 cm in tissue.

Industry Services

Use of nuclear technology applied to research and engineering problems such as quality control, wear rates, corrosion, product integrity, process tracing, etc. Applicable to many industries.

Reactor Technology

Applied nuclear chemistry for engineering testing of reactor concepts.

Biomedical Research

Radioactivity tracing studies for the pharmaceutical, cosmetic, food, chemical, packaging, and other pertinent industries.

Radiation Protection

Complete health and safety programs for all users of radioactivity. Includes: medical facility surveys; soil, vegetation, water, air, milk analyses from reactor sites; routine urinalyses.

Radiation Biology

Research programs to determine the effects of radiation on biological material.

Consulting and Analytical Services

Radioactivity tracing studies in laboratory animals and human subjects.
Autoradiography.
Low level counting.
Dosimetry.
Activation analyses (neutron and charged particles).
Radioactive gas analyses.
Tritium and C^{14} determinations.
Deuterium measurements.
Isotope dilution.
Leukopenic assays.
Reticulo-endothelial studies.
Product sterilization.
Enzyme activity studies.
Biosynthesis.

Products

Accelerator-produced radioisotopes, virtually chemically pure. Laboratory standards and custom sources. Selected reactor-produced radioisotopes. Neutron dosimeters, standard and custom. Custom labeled compounds.

NSEC

Nuclear Science and Engineering Corporation

P. O. Box 10901 • Pittsburgh 36, Pa. • HOmestead 2-4000 • Area Code 412 • TWX PG-940