

99-keV photon. Furthermore, the coincidence rate was in agreement with the assignment of the 99-keV photon to a gamma ray with conversion coefficient ~ 10 .

As will be borne out in the discussion, it is reasonable to assign the 150-keV transition to another of the series of cascading *E2* transitions (see Fig. 1). On this basis, its conversion coefficient would be 2 and the population of the 295-keV level would be 3×10^{-3} percent. This intensity was below the limits of our detection of an alpha group which for this case was $\sim 10^{-2}$ percent, so the failure to observe the corresponding alpha group is not inconsistent with the decay scheme.

IV. DISCUSSION

The energy levels of U²³⁴ observed from the alpha decay of Pu²³⁸ conform in remarkable fashion with the picture for even-even nuclei well removed from a closed shell developed by Bohr and Mottelson.² In this model collective aspects of nuclear structure and individual particle aspects are coupled and in a region well removed from a closed shell there should be a series of energy levels corresponding to a rotational band in which only even spin states (0+, 2+, 4+, ...) appear. On this basis electric quadrupole transitions should predominate. It was seen that the 44-keV transition definitely falls into this category and the 99-keV transition

probably does so. A unique assignment could not be made for the 150-keV gamma ray because the conversion coefficient could not be measured, but the failure to observe an alpha particle leading to the 295-keV state means that the conversion coefficient is not an order of magnitude greater than unity and is therefore in the proper range for an *E2* transition.

Another aspect of the Bohr and Mottelson model concerns the energy level spacings of the states 0, 2, 4, ... The spectrum is that of a rigid rotator although it should be pointed out that in the treatment the moment of inertia is not that of a rigid body but is an effective moment associated with the angular momentum of the surface waves. The rotational energy levels go as $I(I+1)$, where I is the spin number so the energy levels go as 1, 3.3, 7.0 for the first three excited states of spin 2, 4, 6. In this case, the ratio between the second- and first-excited states (145/44) is 3.3 and between the third and first (295/44) is 6.7. This striking agreement with the theory is the principal reason for assigning the second- and third-excited states 4+ and 6+, respectively, in the absence of independent means of making assignments.

We wish to acknowledge the assistance of Mrs. J. A. Simmons in counting the alpha tracks.

Measurement of the Tritium Concentration in Natural Waters by a Diffusion Cloud Chamber*

E. L. FIREMAN AND D. SCHWARZER
Brookhaven National Laboratory, Upton, New York
(Received January 8, 1954)

The radioactivity of tritium is easily recognized and visually counted in a hydrogen-filled diffusion chamber. A 6-in. diameter diffusion chamber, filled with 100 psi of hydrogen obtained by completely converting 14 cc of water in a Mg furnace, is used to measure the tritium concentration in surface water of the Long Island Sound, in New Jersey well water, and in a number of rain and atmospheric moisture samples. These samples had tritium concentration (T/H ratios) that ranged from 10^{-15} to 3×10^{-18} . Some samples were measured directly, others required some electrolysis to give measurable tritium concentrations. No tritium could be detected in glacial water.

INTRODUCTION

THE existence of natural tritium in Norwegian lake water has been demonstrated.¹ This was accomplished by enriching the natural tritium content by electrolysis between 1 and 10 million fold. A series of measurements on natural tritium in rain and ground water is being carried out at the University of Chicago² with a low-level Geiger counter and an electrolytic

plant. We have applied a diffusion chamber technique to the same problem. This technique is more sensitive than a low-level Geiger counter³ that we have been using here. Therefore, considerably less enrichment is required to measure the tritium in natural waters. In fact, in two rain samples and one atmospheric moisture sample the tritium could be measured directly without any enrichment.

DESCRIPTION OF APPARATUS

Figure 1 is a schematic diagram of the diffusion chamber and its associated hydrogen-converting ap-

³ E. L. Fireman, *Phys. Rev.* **91**, 922 (1953).

* Research performed under the auspices of the U. S. Atomic Energy Commission.

¹ Grosse, Johnston, Wolfgang, and Libby, *Science* **113**, 1 (1951).

² W. F. Libby, *Phys. Rev.* **93**, 1337 (1954); *Proc. Natl. Acad. Sci.* **39**, 245 (1953).

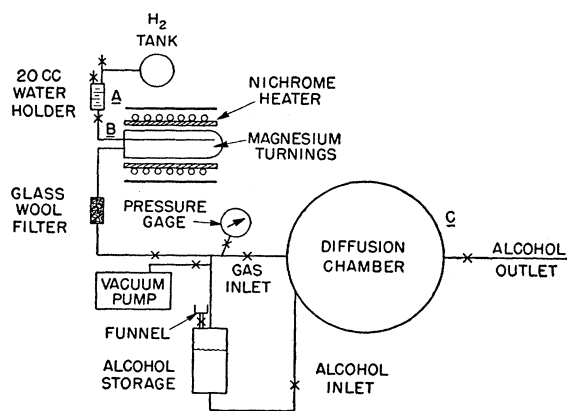


Fig. 1. Schematic block diagram.

paratus. This equipment takes up little space, is easy to build, requires no shielding, requires no electronic equipment and less than 20 amperes from a 110-volt wall plug.

The chamber is filled by putting the water sample into a 20-cc medical syringe, labeled *A* in Fig. 1. Pressure from a standard H_2 tank forces the water into Mg furnace, *B*, which had previously been evacuated and heated to $650^\circ C$. The hydrogen evolved passes through a glass wool filter into the diffusion chamber. A cold trap originally in the line was removed as unnecessary because no water was trapped. Stoichiometry gives a good check on the completeness of the conversion. The complete conversion of 14 cc of water fills the unit to 100-psi pressure. The Mg turnings in the furnace are replaced after four chamber fillings. Figure 2 is a drawing of the Mg furnace.

After a measurement that shows little tritium activity the furnace and cloud chamber are merely evacuated before the next filling. However, if more than 10 tritium counts/minute appear in the chamber it is necessary to convert 5 cc of nonactive water in the furnace and flush the chamber with tank H_2 in order to eliminate traces of tritium.

The conversion of 14 cc of water requires about 15 minutes time. Visual counting can be started 30 minutes later when the chamber reaches temperature equilibrium.

Figure 3 is a cross section of the diffusion chamber. It consists of a 6-in. diameter Pyrex cylinder, 3 in. high and $\frac{1}{4}$ in. thick, between a stainless-steel chamber bottom and steel ring acting as a central temperature control. A 1-in. Bakelite ring insulates

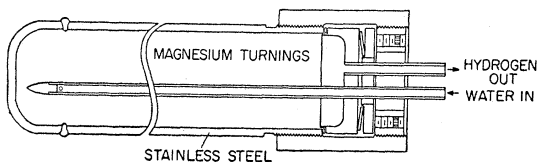


Fig. 2. Magnesium furnace.

the alcohol tray from the central ring. Above this combination of cylinder and rings is the viewing window consisting of a $\frac{1}{2}$ -in. sheet of Allite and a 1-in. sheet of Lucite. The whole thing is clamped together by twelve $\frac{1}{4}$ -in. diameter stainless steel studs.

The chamber bottom is cooled to $-65^\circ C$ by circulating trichlorethylene at dry ice temperature through copper tubing that is soldered to the chamber bottom. Black dye in alcohol on the chamber floor serves as an excellent viewing background. The height of the sensitive layer is almost the same as that of the Pyrex cylinder. The central temperature ring is cooled to $-25^\circ C$ by connecting it to the bottom with 24 copper rods of $\frac{3}{16}$ -in. diameter. A Lucite cylinder surrounds

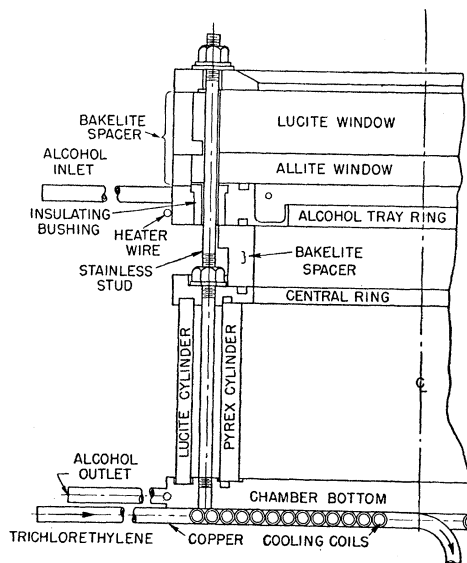


Fig. 3. Diffusion chamber.

the copper rods and Pyrex cylinder and insulates them from the outside. A heater wire about the alcohol tray heats it to $+10^\circ C$. The above temperatures are not very critical; a few degrees variation does not affect the chamber operation.

ELECTROLYSIS

The tritium in some water samples is concentrated by electrolysis. The procedure described by Urey⁴ is followed. Two nickel cells are being used. One concentrates 250 cc of water with 5 g of NaOH to 20 cc. The other concentrates 1500 cc with 30 g of NaOH to 200 cc. A conventional 75-ampere battery charger supplies sufficient current and voltage. The cells are cooled to about $55^\circ F$ with tap water; no provision is made to catch escaping water vapor. The calibration of the tritium enrichment is done with calibrated tritiated water and with rain whose tritium concentration can be measured directly. The fractionation factor

⁴ H. C. Urey and G. K. Teal, *Revs. Modern Phys.* 7, 34 (1935).

under these conditions is (6.9 ± 1.0) , so that the electrolysis of 1500 cc of water to 20 cc increases the tritium concentration by a factor of 40. In the measurements carried out so far, no sample was enriched by more than a factor of 40.

VISUAL COUNTING METHOD

The tritium tracks are short electron tracks with ranges of $\frac{1}{8}$ in. or less in the chamber; many appear as a sharp dot. This type of track is readily distinguished from the extraneous background. Figure 4 is a strip of motion picture film taken of a diffusion chamber with the tritium concentration (T/H ratio) of (1.4×10^{-16}) . The arrows point to the tritium tracks. The appearance of the tritium tracks was ascertained by passing some tank deuterium through a Pd thimble and injecting it into the chamber.

Although tritium can be recorded and counted by flash pictures or by movies, the simplest and most sensitive method is simply to visually observe the tracks as they appear. Different observers without previous training agreed in their counting as well as the statistical error permitted. With a little practice observers can count the tritium tracks occurring within a 6-in. circle. This fact determined the size of the diffusion chamber.

A large part of the data was taken by two independent observers, each without knowledge of the origin of the sample or of the others results.

RESULTS

Table I presents the data. Each measurement is the result of 10 to 20 minutes of separate visual counting by two independent observers. The tritium-like tracks are counted both through an eyepiece limiting the view to a $2\frac{1}{2}$ -in. circle and with unlimited view of the whole chamber. The background through the eyepiece is 0.70 ± 0.06 track/min; in the whole chamber it is 3.26 ± 0.14 tracks/min.

The diffusion chamber is calibrated both with tritiated hydrogen gas and with tritiated water. The tritiated hydrogen gas is tank deuterium ($T/H = 3.6 \times 10^{-12}$) diluted with tank hydrogen to the concentration, ($T/H = 3.6 \times 10^{-16}$). This filling gives 2.50 ± 0.20 counts/min through the eyepiece, and 11.2 ± 0.5 counts/min in the whole chamber. Tritiated water assayed to have a tritium concentration of 9.0×10^{-12} is diluted with New Jersey well water in the ratios of 1 part per 10 000; 1 part per 50 000; 1 part per 250 000, and 1 part per 1.5×10^6 parts of New Jersey well water. Since the New Jersey well water itself was measured to have natural tritium concentration of 3×10^{-18} , its natural tritium raises the most dilute calibration sample from 6×10^{-18} to 9×10^{-18} . The results obtained with these calibrated waters are given in Table I.

The results that have been obtained with various natural waters are also given in Table I. One of the

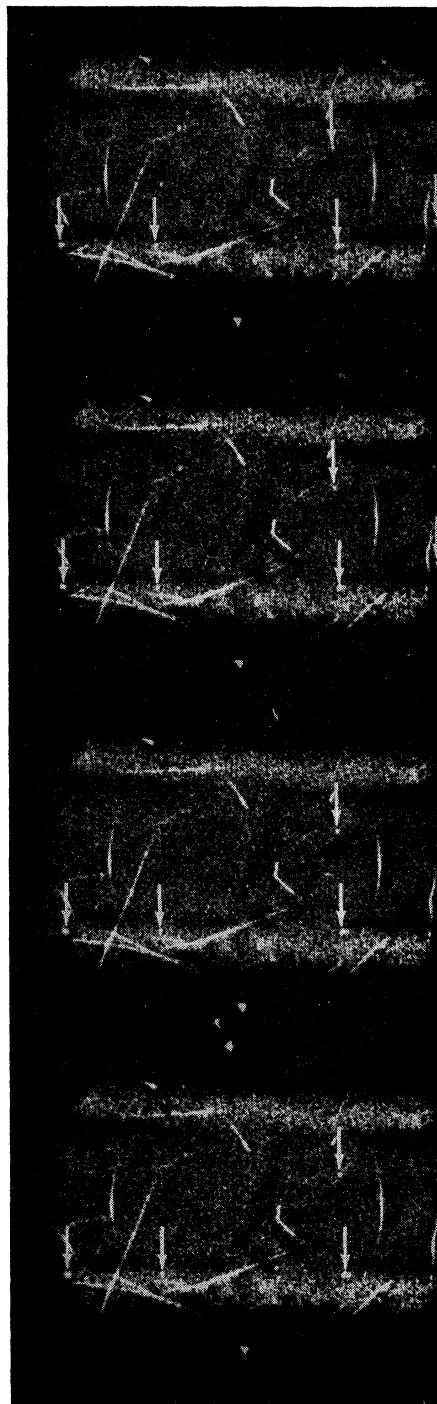


FIG. 4. Strip of 16-mm motion picture film of chamber. (Arrows point to tritium tracks.)

interesting features of the results is the variation of the tritium concentration between different samples, even between different rains there are great differences. The May 26th rain over Long Island had a T/H ratio of 800×10^{-18} , while the August 17th rain over Long

TABLE I. Summary of data.

Sample	View through eyepiece		Enrichment factor	T/H×10 ¹⁸	Whole chamber view		T/H×10 ¹⁸
	Tracks/min	Tracks-background/min			Tracks/min	Tracks-background/min	
8/2/53 Rain	2.45±0.30	1.75±0.30	1	350±60	8.2±0.8	5.0±1.0	225±45
8/2/53 Rain	2.08±0.23	1.38±0.29	1	276±58	9.6±0.8	6.4±1.0	288±45
8/2/53 Rain (electrolyzed)	20.5±1.0	19.8±1.1	14	282±16	100±6	97±6	311±19
8/2/53 Rain (electrolyzed)	3.55±0.30	2.85±0.36	2	285±36	16.90±0.65	13.74±0.8	310±18
Long Island Sound H ₂ O	1.05±0.23	0.35±0.29	1	70±60	3.6±0.8	0.34±1.0	15±45
Long Island Sound H ₂ O (electrolyzed)	0.98±0.16	0.28±0.21	10	5.6±4.0	5.43±0.38	2.17±0.50	9.8±2.3
5/26/53 Rain	4.2±0.3	3.5±0.3	1	700±70	18.2±0.7	15.0±0.8	800±50
7/30/53 Rain	3.68±0.30	1.98±0.40	13	30±10	17.6±1.2	14.3±1.3	49±10
Glacial H ₂ O	0.78±0.14	0.08±0.20	1	<40	3.48±0.28	0.22±0.42	<20
Glacial H ₂ O (electrolyzed)	0.71±0.13	0.01±0.19	40	<1	3.20±0.27	-0.12±0.40	<0.5
Glacial H ₂ O	0.63±0.13	-0.07±0.19	1	<40	2.78±0.26	-0.48±0.40	<20
New Jersey Well H ₂ O	0.75±0.14	0.05±0.20	1	<40	3.50±0.30	0.22±0.44	<20
New Jersey Well H ₂ O	0.55±0.12	-0.15±0.18	1	<40	3.28±0.29	0.02±0.43	<20
New Jersey Well H ₂ O (electrolyzed)	0.80±0.14	0.10±0.20	8	<6	4.05±0.33	0.79±0.47	4.5±2.7
New Jersey Well H ₂ O	0.78±0.14	0.08±0.20	1	<40	3.65±0.30	0.39±0.44	<20
New Jersey Well H ₂ O (electrolyzed)	0.98±0.15	0.28±0.21	16	3.5±2.6	4.20±0.33	0.94±0.47	2.7±1.4
11/27/53 Moisture	1.05±0.16	0.35±0.22	1	70±45	4.25±0.33	0.99±0.47	45±21
11/24/53 Moisture	0.65±0.13	-0.05±0.19	1	<40	2.88±0.27	-0.38±0.40	<20
10/12/53 Rain	1.23±0.18	0.53±0.24	2.5	40±20	5.55±0.38	2.29±0.52	41±10
8/17/53 Rain	1.85±0.20	1.05±0.26	13	16±5	8.08±0.45	4.82±0.59	16.9±4
H ³ Calibration	1.60±0.20	0.90±0.26	1	180	7.36±0.43	4.14±0.49	180
H ³ Calibration	5.45±0.38	4.75±0.44	1	900	20.1±0.7	16.9±0.8	900
H ³ Calibration (electrolyzed)	2.28±0.24	1.58±0.30	40	9	10.78±0.53	7.52±0.65	9
H ³ Calibration (electrolyzed)	2.40±0.25	1.70±0.31	8	39	9.4±0.5	6.0±0.6	39

Island had a T/H ratio of 16×10^{-18} . Some particular rains may be affected by bomb blasts; however, the majority of rains and the world reservoir of natural tritium will not be affected by such small-scale events. The variation of tritium in most rains is dependent on the origin and recent history of the rain water. Rain consisting of water evaporated from the continents, or water that has spent much time at a high altitude should have appreciably more tritium than rains composed of recently evaporated ocean water. A discussion of the tritium production in the atmosphere

and its relation to the world reservoir and distribution of natural tritium has been given by one of the authors.³

Water from the Mendenhall glacier⁵ was the only water in which the tritium could not be detected with a factor of 40 electrolytic enrichment, indicating a tritium concentration of less than 5×10^{-19} .

The authors would like to thank Dr. Ralph Shutt for several helpful discussions on diffusion cloud chambers.

⁵ The authors wish to thank Dr. Baden of the Snow, Ice, and Permafrost Research Establishment for this sample.

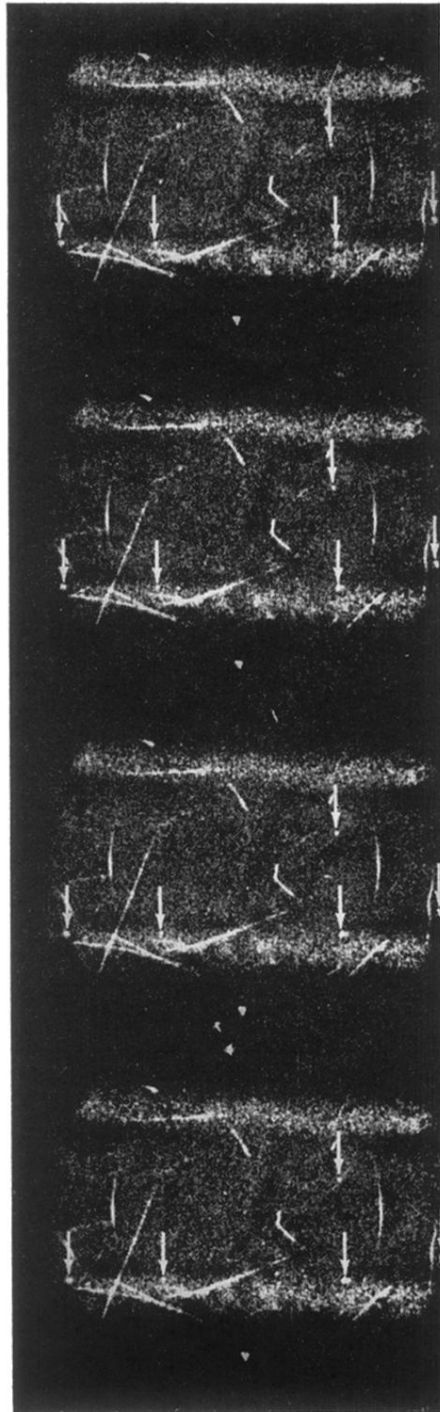


FIG. 4. Strip of 16-mm motion picture film of chamber. (Arrows point to tritium tracks.)