

The Half-Life of Tritium by Absolute Counting*

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Results of a measurement of the half-life of tritium by absolute counting are presented. The method consisted of the mass spectrometric analysis of a hydrogen-tritium mixture enriched in tritium, the dilution of some of this material with hydrogen in a known way to a countable level of activity, and the counting of a portion of the diluted gas in a Geiger counter of known effective volume. The half-life was found to be 12.41 with uncertainties of plus 0.15 and minus 0.25 year.

ANALYSIS, DILUTION, AND MEASUREMENT OF THE TRITIUM

THE initial hydrogen-tritium mixture, enriched in tritium, came from a system of Hertz pumps which has been set up in this Laboratory by Dunn, Mosley, Potter, and Robinson. Unfortunately, it was not possible to utilize this material until some time after it became available, and it became slightly contaminated with hydrogen during this interval. The sample was freed from substances other than hydrogen by forming uranium hydride with well-outgassed and finely divided uranium and then pumping off residual gases. This procedure was repeated several times. The tritium was finally evolved into a Pyrex bulb. This gas was immediately analyzed with a mass spectrometer, a portion subjected to a first-stage dilution with pure hydrogen, and the remaining enriched material again analyzed.

The enriched material was analyzed on a 60° Nier-type mass spectrometer. The various species were brought to focus by varying the magnetic field, and the instrument was operated without the source magnet. The gas was admitted to the spectrometer through a molecular leak. The analyses made just before and just after the first-stage dilution gave 98.17 and 98.21 atomic percent tritium, respectively. A preliminary analysis of a synthetic hydrogen-tritium mixture, of known composition showed that the mass 2 to mass 4 ratio is not in error by more than about two percent due to discrimination effects. A similar conclusion was later reached with a hydrogen-deuterium mixture of known composition.

Preliminary experiments had been made in regard to the effect of the tritium radioactivity on stopcock grease in introducing contamination by formation of hydrogen and other gases, principally methane. In these experiments the situation normally encountered was intentionally greatly magnified and indicated that undesirable effects would be completely negligible for the few hours which elapsed between the final purification of the tritium and the first dilution stage. The diluted gas was protected from contact with stopcock grease by means of mercury.

The dilution of the tritium with pure hydrogen was carried out in three stages in order to reduce the activity to a suitable level. At each stage a known amount of the gas richer in tritium, contained in a volume of about 2 cc, was allowed to mix with a known amount of pure hydrogen contained in a bulb of about 500-cc capacity. Both volumes had been accurately calibrated with mercury. In the determination of the amounts of gases, temperatures were estimated to 0.01°C. In the pressure measurements meniscus positions were estimated to 0.001-cm Hg, appropriate reductions to international centimeters of Hg were made, and small corrections were made for non-ideal gas behavior. The pure hydrogen was present at a higher pressure so that some mixing occurred on connecting the two vessels. In addition, the larger vessel could be cooled with liquid nitrogen to facilitate mixing. At each stage the cycle of cooling and warming was repeated ten times, and as a final precaution the gas was allowed to stand overnight. It was necessary to insure that contamination from a previous stage did not falsify the dilution procedure. For this reason the first-stage dilution was made in a different vacuum system from the remaining two stages, since it was thought that contamination would not be removable to a degree suitable for the last stage. Decontamination consisted in passing hydrogen or water through the apparatus for many hours. For the final stage of dilution new standard volumes were calibrated in order to remove any uncertainty with regard to contamination. Effectiveness of decontamination was checked by the counting of hydrogen gas which had stood for several hours in contact with the apparatus.

Pressure and temperature measurements on some diluted gas in the small calibrated volume mentioned were used to determine the amount of the gas to be transferred to a counter. In order to check on how well gas was transferred, three different systems, presenting much different surface areas, were employed. The first two systems involved Toepler pumps which differed by a factor of eight in volume. In the third arrangement the gas was allowed to distribute itself between the counter, the small calibrated volume, and a small additional connecting volume of capillary tubing. Since about 98 percent of the gas went into the counter and since all volumes involved were known, the amount of

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gas in the counter was accurately known. There seemed to be no systematic differences in the half-life determinations using the different methods of transfer. The transfer systems were pretreated with hydrogen and with some of the diluted tritium gas, since some preliminary results on the untreated glass of the transfer system presenting the largest surface had indicated that a small amount of activity (not more than about $\frac{1}{2}$ percent of the material used in a counting measurement) was assumed by the glass. Apparently, however, an approximately stationary state of saturation was reached by the surface during the course of the experiments reported here, since the effects of prolonged pumping or exposure to D_2 for a few hours did not give half-life values differing in a consistent way from values obtained without so treating the transfer system.

COUNTERS AND COUNTING MEASUREMENTS

Two counters of identical construction were used during the experiments with no difference in results. The counters were about 15 inches long, and their construction has been previously described.¹ The partial pressure of the hydrogen-tritium mixture in the counter was slightly over 1-mm Hg, and to this 93 percent argon-7 percent ethyl alcohol quencher was added in such a way that no tritium escaped. The final pressure was usually 10-cm Hg. The counting rate due to tritium was usually about 45 counts per second. Dead time corrections were in all cases less than 1.5 percent and were made on the basis of a measured dead time of 3.0×10^{-4} sec. Corrections were made for background, which was usually monitored by means of a third similar counter with which the main counters were intercompared periodically.

It was necessary to find the fraction of the disintegrations which gave rise to counts. This may be taken as the ratio of an effective volume of the counter to the total volume occupied by gas. The effective volume may be regarded in first approximation as being bounded by the cylinder wall and by planes which are perpendicular to the counter axis at the points where the central wire emerges from its shields. The length of unshielded wire L should be replaced, however, by $L - \epsilon$, where ϵ is determined by such factors as the nature of the electric field, the geometry of the counter ends, the counter filling, and the particle range. If the counter contains a radioactive gas of decay constant λ present at a density of ρ nuclei per unit volume in a counter of effective cross section A , then the counting rate for a counter of wire length L will be

$$R_L = A\rho\lambda(L - \epsilon). \quad (1)$$

By simultaneously filling two counters, identical except for their wire lengths L , with a mixture of quencher and diluted tritium, ϵ was found to be 0.9_8 and 1.0_8 cm at quencher pressures of approximately 10

and 20 cm Hg, respectively. The longer effective lengths of the same type counters for the gas counting of carbon fourteen¹ are probably due to a greater contribution from the counter ends by the longer range C^{14} β -particles. It is shown below that the effective cross section of the counter is the same as the geometrical cross section to within experimental errors. The use of counters of different dimensions in absolute gas counting appears to be due to Libby *et al.*² and has also been used by others.^{1,3-5}

The counters were sometimes, but not always, allowed to stand in a hydrogen atmosphere to minimize possible adsorption effects. The presence of the tritium in the counter for the exposure time of an experiment did not appear to raise backgrounds. This observation does not preclude the possibility of adsorption during a measurement. Some experiments were made which might have uncovered such an effect. In experiment 10, extra hydrogen equal to that normally present was added with no effect. In addition, the higher pressure of quencher in experiment 5 and the added alcohol in experiment 13 did not show any effect, although these experiments were primarily performed for other reasons (see below). In addition, one of the two counters was considerably older, had been more often used than the other, and probably had a surface more susceptible to adsorption. However, the same half-life is given by both counters within experimental errors, suggesting that loss of counts by surface adsorption of material does not constitute a serious error. Gas adsorbed on the wall might still contribute to the counting rate, since even those β -particles which are directed toward the wall rather than the volume of the counter may be back-scattered or cause the emission of secondary electrons. Any serious effects due to slow absorption are unlikely, since the counting rate was constant to within experimental error over a 2-day period in a preliminary experiment. In experiment 5 the quencher was increased to 20-cm Hg in order to see whether any significant number of β -particles had failed to produce a count at the lower pressure because of their motion toward the wall after having been produced at a distance from the wall of the order of or less than a mean free path for ion pair formation. No such effect was observed, probably because of backscattering, secondary emission, and the short mean free path for the low energy tritium β -particles. The effective and geometrical cross sections of the counter are thus closely the same. It was considered possible, although not likely, that the absence of an effect in experiment 5 might be owing to a balancing decrease in counting rate due to greater electron attach-

² Anderson, Libby, Weinhouse, Reid, and Grosse, *Phys. Rev.* **72**, 931 (1947).

³ Hawkins, Hunter, Mann, and Stevens, *Phys. Rev.* **74**, 696 (1948).

⁴ W. B. Mann and G. B. Parkinson, *Rev. Sci. Instr.* **20**, 41 (1941).

⁵ Engelkemeier, Hamill, Inghram, and Libby, *Phys. Rev.* **75**, 1825 (1949).

¹ W. M. Jones, *Phys. Rev.* **76**, 885 (1949).

ment or recombination at the higher pressure. Since argon and hydrogen give no trouble with electron attachment, while the alcohol might, the amount of alcohol present in experiment 13 was made more than twice that normally present with no indication of an effect. In experiment 17 a small amount of air was added without effect on the half-life obtained, while a mass spectrometric analysis of quencher gas from which the alcohol had been frozen out showed the oxygen content to be less than 0.01 percent. Thus, electron attachment does not seem to be an important factor. Recombination of the electrons and positive ions should be negligible at the pressures prevailing in the counter. In experiment 12 the 0.005-in. Kovar wire was replaced by one of wolfram on the chance that some take-up or evolution of gas during a counter discharge might be occurring, but such behavior does not appear to be significant.

The plateaus were definite, but their length was considerably shortened by the presence of the hydrogen. An experiment was therefore performed to verify that the operating voltage had been properly chosen and that the hydrogen was not causing any additional effect on counting rate. A uranium foil was taped to the counter and counted with hydrogen plus quencher in the counter and with quencher alone, in which case a much longer plateau region was obtained. Several counting rate measurements were taken in each situation, and the ratio of the counting rate with H_2 to that without H_2 was found to be 1.003 ± 0.004 . Two different high voltage supply-scaler circuits were used with no apparent difference in results, and no detectable change in counting rate was found by use of a shorter cable to the counter. A small correction to the half-lives, amounting to a little over 1 percent in the last experiments, was made to allow for tritium decay subsequent to the time of analysis.

DISCUSSION OF ERRORS AND COMPARISON WITH OTHER WORK

The probable error from the data of Table I is 0.04 year. There is a negligible uncertainty associated with the mass spectrometric analysis, the errors from measurements in the three stages of dilution may correspond to 0.01 yr, and the fraction of the disintegrations counted, as determined by measurement of ϵ , contributes an estimated uncertainty of 0.02 yr. Uncertainties of 0.01 yr and 0.02 yr may arise from the dead time and background corrections. On this basis a

TABLE I. Summary of experiments.

Experiment	Half-life, years	Experiment	Half-life, years
1	12.41	10	12.32
2	12.37	11	12.44
3	12.49	12	12.50
4	12.42	13	12.32
5	12.38	14	12.42
6	12.34	15	(counter developed leak)
7	12.38	16	12.52
8	12.30	17	12.47
9	12.38		
		Average	12.41 years
		pe	0.04 year

probable error of about 0.05 yr is estimated. Because of uncertainty in each of the experiments performed to uncover sources of absolute error and because of unknown errors, a larger estimate of uncertainty seems appropriate. The half-life of tritium, as given by this work, is taken to be 12.41 years with uncertainties of plus 0.15 and minus 0.25 year.

The above value may be compared with the three most recent published values of the half-life. These values are in agreement within their stated limits of error.

The decay of the current from an ionization chamber containing tritium gas was used to obtain a value of 10.7 ± 2 years.⁶ The precision uncertainty was much lower than the error quoted, which was stated to arise from uncertainties due to possible tritium absorption and to the effect of the He^3 produced.

The production of He^3 from two samples of tritium gas over a period of a few months gave a value of 12.1 ± 0.5 years.⁷

Measurement of the steady-state rate of diffusion of He^3 from a thin-walled quartz capsule containing a known amount of tritium as water gave a value of 12.46 ± 0.2 years.⁸ In a subsequent and more detailed account⁹ the uncertainty was given as 0.1 year.

A precise determination of the tritium half-life by measurement of the He^3 growth has been in progress for some time and will be reported soon.

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⁶ Goldblatt, Robinson, and Spence, Phys. Rev. **72**, 974 L (1947).

⁷ A. Novick, Phys. Rev. **72**, 972 L (1947).

⁸ Jenks, Ghormley, and Sweeton, Phys. Rev. **75**, 701 L (1949).

⁹ Jenks, Sweeton, and Ghormley, Phys. Rev. **80**, 990 (1950).