

Matematisk-fysiske Meddelelser  
udgivet af  
Det Kongelige Danske Videnskabernes Selskab  
Bind **35**, nr. 8

---

Mat. Fys. Medd. Dan. Vid. Selsk. **35**, no. 8 (1966)

---

# SOME ACCURATE HALF-LIFE DETERMINATIONS

BY

V. MIDDELBOE



København 1966  
Kommissionær: Munksgaard

## Synopsis

The rate of decay of three short-lived nuclides has been re-investigated and improved half-life values have been found. The results are as follows:  $\text{Ba}^{137\text{m}}$   $153.46 \pm 0.20$  s,  $\text{Ag}^{109\text{m}}$   $39.80 \pm 0.18$  s, and  $\text{Rh}^{106}$   $30.35 \pm 0.15$  s. The standard deviations include the uncertainties involved in the investigations of radioactive impurities.

Some measurements in 1960–61 by the author on the half-life of  $\text{Ba}^{137\text{m}}$  seemed to indicate that the current standard value,  $2.60 \pm 0.05$  minutes, is about two per cent too high, and through personal communication the following comment on some below-mentioned biological work was received: "... our half-lives seem to run about 2.5 minutes from those tissues which gave the best measurements. However, we do not feel that these measurements are particularly precise, ..."

A meticulous re-determination of the half-life of  $\text{Ba}^{137\text{m}}$  was then commenced, serious consideration being given to objective evaluation of impurity bias and natural uncertainty. Improved values for the half-lives of  $\text{Ag}^{109\text{m}}$  and  $\text{Rh}^{106}$  were subsequently obtained by applying the methodology developed for  $\text{Ba}^{137\text{m}}$ .

## Introduction

Three short-lived radioactive nuclides have been re-investigated and in each case a more accurate half-life value has been determined. The test nuclides were selected with a view to objective evaluation of the radio-purity of the sample and the natural uncertainty of the result.

In principle, a half-life value is found by isolating a sample of the test nuclide and measuring its rate of decay. However, the result may be biased by a radioactive impurity in the sample, and this type of uncertainty is hard to evaluate. On the other hand, the uncertainty of the result due to the random nature of radioactive decay is easy to evaluate when the well-known ratio method of half-life measurement is applicable.

In 1959, it was found that the half-life of biologically separated  $\text{Ba}^{137\text{m}}$  was slightly smaller than the value currently accepted by physicists. This apparent discrepancy could perhaps have been due to a difference in the radio-purity of the differently separated samples. Certainly, a better standard value for the physical half-life of  $\text{Ba}^{137\text{m}}$  is needed, since agreement between the observed and the standard  $t_{1/2}$  value is used as a means of checking consistency in the type of biological work referred to here<sup>(1)</sup>.

## Methodology

### Method of Measurement

Since the half-lives of the selected nuclides range from 0.5 to 2.5 minutes, the ratio method of measurement is practicable. In this method the investigator observes the ratio between the amount of test nuclide remaining in a sample after a period of decay and the amount initially present. Below, the following inverse ratio, the fall-off factor, is used:

$$F = \frac{N_0}{N_T}$$

where  $N_0$  = number of test nuclei at zero time

$N_T$  = number of test nuclei  $T$  minutes later.

When an  $F, T$  set has been observed, the corresponding value for the half-life of the test nuclide is found by use of the equation

$$t_{\frac{1}{2}} = T \frac{\ln 2}{\ln F}. \quad (1)$$

Equation (1) is a simple transcription of Rutherford's law,  $-dN = \lambda N dt$ , in the integrated form  $\ln F = \lambda T$ .

A fall-off factor is measured in the following way. A sample of test nuclide is placed near a radiation detector and pulse counting is started simultaneously with timing. The counts are summed continuously throughout a period of time,  $\theta$ , of sufficient duration for virtually all the test nuclei to decay, and an observation is made of the total sum-count,  $\Sigma'$ , thus accumulated. Also, an observation of the sum-count,  $S'$ , and the corresponding counting time,  $T$ , is made (without stopping the counter) fairly early in the course of the total period of time,  $\theta$ . Finally, the background radiation is assayed by counting the decayed sample for a period of time, for instance equal to  $\theta$ , and observing the background sum-count,  $B$ , thus accumulated.

The value of the fall-off factor, that corresponds to the observations described above, is found by use of the simple equation

$$F = \frac{\Sigma}{\Sigma - S} \quad (2)$$

where  $\Sigma = \Sigma' - B$  counts

$$S = S' - B \frac{T}{\theta} \text{ counts.}$$

Equation (2) rests on the assumption, that the counting efficiency remains constant throughout the entire  $2\theta$  period of observation.

### Natural Uncertainty

The statistical fluctuation of replicate  $t_{\frac{1}{2}}$  values is mainly due to the natural randomness inherent in the emission and absorption of radiation. When the ratio method of  $t_{\frac{1}{2}}$  measurement is used, the natural standard deviation of the observed  $t_{\frac{1}{2}}$  value can be closely approximated by the expression

$$\frac{\sigma_{\text{nat}}[t_{\frac{1}{2}}]}{t_{\frac{1}{2}}} = \frac{1}{\sqrt{\Sigma \ln F}} \sqrt{F-1 + 2(F-1)^2 \beta / \Sigma} \quad (3)$$

where  $\beta = B(\theta - T)/\theta$ .

If  $\beta$  is completely insignificant in comparison with  $\Sigma$ , equation (3) indicates that  $\sigma_{\text{nat}}[t_{\frac{1}{2}}]$  is minimal for  $F = 5$ , which is identical to the finding of PEIERLS<sup>(2)</sup>. If  $\beta$  is, say, one tenth of  $\Sigma$ , then  $F = 3$  actually minimizes  $\sigma_{\text{nat}}[t_{\frac{1}{2}}]$  according to equation (3), but  $\sigma_{\text{nat}}[t_{\frac{1}{2}}]$  is still only ten per cent above minimum for  $F = 5$ .

For typical experimental values such as  $\beta = 10^4$  counts,  $\Sigma = 10^5$  counts and  $F = 4$ , equation (3) gives  $\sigma_{\text{nat}}[t_{\frac{1}{2}}] = 0.005 \cdot t_{\frac{1}{2}}$ , in which case the natural standard deviation of the mean of 25 replicate measurements is one part in a thousand.

### Dead-Time Correction

Loss of counts at high counting rates tends to bias the observed half-life value upwards. Using the available apparatus and a fixed  $F$  value, the observed  $t_{\frac{1}{2}}$  should be expected to increase linearly with the initial counting rate,  $R_0$ , and this is also in agreement with experimental observations up to almost 200 kc/min. (kilocounts per minute). Consequently, any dead-time bias on  $t_{\frac{1}{2}}$  is eliminated conveniently by linear extrapolation of  $t_{\frac{1}{2}}$  as a function of  $R_0$  to the point at which  $R_0 = 0$ .

The dead-time loss on the counting rate,  $R$ , at any moment,  $t$ , may be theoretically approximated by

$$- \tau R^2 = - \tau R_0^2 \cdot e^{-2\lambda t}$$

where  $\tau$  = dead-time.

From which we find (assuming the initial rate is less than 200 kc/min.)

$$\text{bias}_{\tau}[\Sigma] = -\frac{1}{2} \tau R_0 \Sigma, \quad \text{and} \quad \text{bias}_{\tau}[\Sigma - S] = -\frac{1}{2} \tau R_T (\Sigma - S)$$

so, since  $F = \Sigma/(\Sigma - S)$ , and considering equation (1),

$$\frac{\text{bias}_{\tau}[F]}{F} = -\frac{1}{2} \tau (R_0 - R_T), \quad \text{and} \quad \frac{\text{bias}_{\tau}[t_{\frac{1}{2}}]}{t_{\frac{1}{2}}} = \frac{1}{2} \tau (R_0 - R_T) / \ln F.$$

Finally, since  $R_T = R_0/F$ , we may write

$$\frac{\text{bias}_{\tau}[t_{\frac{1}{2}}]}{t_{\frac{1}{2}}} = \frac{1}{2} \tau \frac{F-1}{F \cdot \ln F} R_0. \quad (4)$$

### Time-Keeping

The author measures the critical period of time,  $T$ , by an eye-and-hand method, i.e., a stop-watch is started at counting time zero and stopped by hand as the eye sees the sum-count pass through an appropriate round number. This operation is analogous to timing a runner in athletics, which is known to be reproducible to 0.1 second (or 0.002 minute) by a trained time-keeper.

When all the timing is done by one and the same investigator, the possibility of a systematic error in timing becomes serious. The following control experiment was therefore designed. Three determinations of the half-life of  $\text{Ba}^{137\text{m}}$  are carried out with  $T$  equal to 0.5, 6.5 and 16 minutes, respectively, and  $R_0$  equal to 40, 80 and 160 kc/min., respectively. The dead-time bias on the half-life value is then the same in each of the three determinations (cf. eq. (4) and Table 1). However, a small systematic error in timing must bias the first half-life determination relatively strongly and the other two only slightly.

TABLE 1. Data for control experiment on time-keeping.  $F = 2^{T/t_{1/2}}$ .

$T$ $t_{1/2}$	$F - 1$ $F \cdot \ln F$	$R_0$ (rel.)
0.19	0.94	1
2.5	0.47	2
6	0.24	4

Each of the three determinations comprised 25 single half-life measurements and the mean values obtained were  $2.558 \pm 0.004$ ,  $2.560 \pm 0.001$  and  $2.560 \pm 0.002$  minutes, the stated figure of uncertainty being in each case the root-mean-square deviation

$$\sigma_{\text{rms}}[\bar{t}_{1/2}] = \sqrt{\frac{\sum(t_{1/2} - \bar{t}_{1/2})^2}{n(n-1)}}$$

where  $t_{1/2}$  = single observation

$\bar{t}_{1/2}$  = mean observation

$n$  = number of replicates.

The  $\bar{t}_{1/2}$  value obtained with  $T = 0.5$  minute, namely  $2.558 \pm 0.004$ , is apparently unbiased in comparison with the other two  $\bar{t}_{1/2}$  values. This indicates a null systematic timing error with an uncertainty of about  $\pm 0.001$

minute, since a systematic timing error of 0.001 minute on  $T$  would bias the 2.558-minute value by 0.005 minute (cf. eq. (1)), which is of the same magnitude as the  $\sigma_{\text{rms}}$  of this value and would therefore not normally be detected by the control experiment performed.

Thus we may conclude, that under the prevailing conditions of measurement the uncertainty due to the possibility of a systematic error in timing, (below the sensitivity of the control experiment) contributes in general

$$\left(\frac{t_{\frac{1}{2}}}{T} 0.001 \text{ minute}\right)^2 \quad (5)$$

to the overall variance of  $\bar{t}_{\frac{1}{2}}$ .

### Radio-Purity Investigation

Consider two samples, identical in test nuclide content, which contain an alien radio-nuclide in the relative amounts  $X:1$ . To a first order approximation the impurity bias on the one sample's  $t_{\frac{1}{2}}$  value is then  $X$  times that on the other. Consequently, the sum of the two impurity biases and the difference between them stand in the ratio of  $(X+1):(X-1)$ , and the following equation is applicable:

$$\frac{\bar{b}^*}{\frac{1}{2} \Delta} = \frac{X+1}{X-1} \quad (6)$$

where  $\bar{b}^*$  = impurity bias on the mean of the two  $t_{\frac{1}{2}}$  values

$\Delta$  = difference between the two  $t_{\frac{1}{2}}$  values.

Two samples possessing the above properties can be produced experimentally as follows. Take one sample, the measurement of which is started immediately, and another sample composed of  $2^N$  of the first kind, the measurement of which is started after a delay of  $N \cdot t_{\frac{1}{2}}$  minutes. In comparison with the immediate content of radio-impurity in the first sample, the content in the second sample at the end of the delay period is different by a factor of

$$X = 2^{N-N^*} \quad (7)$$

where

$$N^* = (N \cdot t_{\frac{1}{2}}) / t_{\frac{1}{2}}^{\#} \quad (8)$$

$t_{\frac{1}{2}}^{\#}$  representing the half-life of the alien radio-nuclide.

Half-life measurements based on the two types of sample described above may be termed immediate and delayed, respectively. When the average difference,  $\Delta$ , between  $n_1$  immediate and  $n_2$  delayed measurements is found to be less than or about equal to the root-mean-square deviation of this difference,  $\sigma_{\text{rms}}[\Delta]$ , then no significant impurity has been detected and the measured  $t_{\frac{1}{2}}$  values possess a null impurity bias. The best result obtainable in this case is the mean,  $\bar{t}_{\frac{1}{2}}$ , of all  $n_1 + n_2$  measurements, but this result is of course no more reliable than the null value of its impurity bias,  $\bar{b}^*$ , of which the uncertainty is (cf. eqq. (6) and (7))

$$\sigma_{\text{rms}}[\bar{b}^*] = \left| \frac{2^{N-N^*} + 1}{2^{N-N^*} - 1} \right| \frac{1}{2} \sigma_{\text{rms}}[\Delta]. \quad (9)$$

Equation (9) shows clearly that the above-mentioned delay method of checking radio-purity is most uncertain when  $t_{\frac{1}{2}}^{**}$  is close to  $t_{\frac{1}{2}}$  (i.e.,  $N^*$  close to  $N$ ). Fortunately, however, the amount of impurity required to induce a significant bias on the observed half-life value increases rapidly as  $t_{\frac{1}{2}}^{**}$  approaches  $t_{\frac{1}{2}}$ , which means that the delay method can be supplemented by a direct search for impurities with half-lives near that of the test nuclide.

Arbitrarily, we will restrict  $\sigma_{\text{rms}}[\bar{b}^*]$  to  $1.5 \cdot \sigma_{\text{rms}}[\Delta]$ . The lower limit of  $|N - N^*|$  is then 1 (cf. eq. (9)) and the half-lives of the potential impurities, that we will attempt to check by the delay method, are restricted to the following "wing-intervals" (cf. eq. (8)):

$$t_{\frac{1}{2}}^{**} \leq \frac{N}{N+1} t_{\frac{1}{2}} \quad \text{and} \quad t_{\frac{1}{2}}^{**} \geq \frac{N}{N-1} t_{\frac{1}{2}}. \quad (10)$$

In other words, the delay method is considered unacceptable for investigating potential impurities belonging to the "centre-interval"

$$\frac{N}{N+1} t_{\frac{1}{2}} < t_{\frac{1}{2}}^{**} < \frac{N}{N-1} t_{\frac{1}{2}}. \quad (11)$$

It is worth noting, that a null  $\Delta$  value conveys sample purity with respect to any number or combination of (genetically unrelated) impurities belonging to either or both wing-intervals (10), because the apparent half-life of a sample containing a radio-impurity increases with time irrespective of whether  $t_{\frac{1}{2}}^{**}$  is greater or less than  $t_{\frac{1}{2}}$ , and two or more (independent)  $\Delta$  contributions will therefore always enhance one another.



Let the fall-off factor,  $2^N$ , of the test nuclide during the delay period be, e.g., six. The centre-interval (11) is then given by  $0.7 < t_{\frac{1}{2}}^*/t_{\frac{1}{2}} < 1.7$  and, in principle, every radio-nuclide that satisfies this condition must be looked for directly in samples of the test nuclide. Table 2 indicates the relative amount of any such nuclide required to induce a bias of one part in a thousand on the apparent half-life of the test nuclide (when  $F$  is also 6).

The relative bias on an observed  $t_{\frac{1}{2}}$  due to an alien radio-nuclide,  $c^*$ , belonging to the centre-interval  $0.7 < t_{\frac{1}{2}}^*/t_{\frac{1}{2}} < 1.7$ , is

$$\frac{\text{bias}_{c^*}[t_{\frac{1}{2}}]}{t_{\frac{1}{2}}} = \frac{F^{1-t_{\frac{1}{2}}^*/t_{\frac{1}{2}}} - 1}{\ln F} \cdot \frac{\Sigma^*}{\Sigma} \tag{12}$$

where  $\Sigma^*$  = sum-count of impurity at counting time  $\theta$ .

TABLE 2. The percentage of centre-interval impurity that biases the half-life of the test nuclide by one part in a thousand, when  $F = 6$ . Calculated by use of equation (12).

BIAS OF -1 PER MILLE ON $t_{\frac{1}{2}}$		BIAS OF +1 PER MILLE ON $t_{\frac{1}{2}}$	
$t_{\frac{1}{2}}^*/t_{\frac{1}{2}}$	$\Sigma^*/\Sigma$ ‰	$t_{\frac{1}{2}}^*/t_{\frac{1}{2}}$	$\Sigma^*/\Sigma$ ‰
0.7	0.3	1.1	1.0
0.75	0.4	1.2	0.5
0.8	0.5	1.3	0.4
0.85	0.6	1.4	0.3
0.9	0.9	1.5	0.2
—	—	1.7	0.2

Finally, the presence of an unfortunate combination of genetically related and unrelated impurities might bias  $t_{\frac{1}{2}}$  significantly and yet produce a zero  $\Delta$  value. However, in the work presented here the only nuclides that could conceivably be the cause of such a chain effect are given in Table 3.

TABLE 3. Genetically related nuclides, two or more of which might contribute to a significant chain effect.

Ra-224	Th-234	Rn-222	Th-227	Ra-225
Rn-220	Pa-234m	Po-218	Ra-223	Ac-225
Pb-212		Pb-214	Rn-219	Fr-221
Bi-212		Bi-214	Pb-211	Bi-213
Tl-208			Bi-211	Tl-209
			Tl-207	Pb-209

A special case of genetical contamination is of course the presence of some of the test nuclide's parent in the sample. This does not bias the half-life of the sample significantly when the parent nuclide is very long-lived relative to the daughter, but it does increase the radiation background,  $B$ , and thereby the natural uncertainty of the observed half-life value as expressed in equation (3).

## Barium 137m

### Previous Results

Several nuclear properties of  $\text{Ba}^{137\text{m}}$  were published in 1949 by MITCHELL and PEACOCK<sup>(3)</sup>. Their half-life value was  $156 \pm 3$  sec. which corresponds to  $2.60 \pm 0.05$  min.

In 1948, TOWNSEND, CLELAND and HUGHES<sup>(4)</sup> found the value  $2.63 \pm 0.08$  min. They added sulphate ions to a solution containing  $\text{Cs}^{137}\text{-Ba}^{137\text{m}}$ . This solution was brought into contact with a piece of solid  $\text{BaSO}_4$  which was subsequently washed, dried and counted. The initial counting rate was 12 kc/min.

Also in 1948, ENGELKEMEIER<sup>(5)</sup> reported the value 2.5 min.

### Separation Technique

$\text{Ba}^{137\text{m}}$  is easily separated from  $\text{Cs}^{137}$  by elution with basic EDTA from  $\text{Cs}^{137}$  absorbed on a conventional cation exchange column. The total number,  $\Sigma$ , of  $\text{Ba}^{137\text{m}}$  counts in the sample can be pre-determined by counting the drops of eluate included in the sample.

### Present Result

240 half-life measurements were made (with  $F = 6$ ) using a NaI crystal and counting all the pulses above the input bias voltage. The main values that were found are given in Table 4.

The 110 measurements at  $R_0 = 27$  kc/min. comprise 55 immediate measurements averaging  $2.558_5 \pm 0.001_5$  min. and 55 delayed measurements (with  $2^N = 6$ ) averaging  $2.558_0 \pm 0.001_5$  min. The difference,  $\Delta$ , is nil and, consequently, no wing-interval impurity was present in the samples. The uncertainty of this conclusion imposes  $\pm 1.5 \cdot \sigma_{\text{rms}}[\Delta] = \pm 0.0032$  min. on the mean of the 110 measurements (see page 8); and the extrapolated value of  $t_{1/2}$  must also carry this standard deviation.

TABLE 4. 240 measurements of the half-life of Ba<sup>137m</sup>.

Number of replicates ( <i>n</i> )	Initial count-rate ( <i>R</i> <sub>0</sub> ) kc/min.	Mean value observed ( $\bar{t}_{\frac{1}{2}}$ ) min.	Precision of mean value ( $\sigma_{\text{rms}}[\bar{t}_{\frac{1}{2}}]$ ) min.
55	160	2.559	0.001
50	80	2.560	0.001
25	40	2.558	0.002
110	27	2.558	0.001
	( 0	2.558*	0.001 )

\* by extrapolation

A previous determination of the half-life of Ba<sup>137m</sup> had been made using a well-crystal detector which was connected to a scaler with a dead-time of about 10<sup>-7</sup> min. 110 measurements were carried out utilizing *F* values which were proportional to *R*<sub>0</sub>, so that the counting rate at the stop watch moment, *T*, was the same in all the measurements. However, a variation in the dead-time bias of the measured *t*<sub>½</sub> values was attained (cf. eq. (4)), making it possible to extrapolate to zero dead-time bias. The main values that were found are given in Table 5.

TABLE 5. 110 measurements of the half-life of Ba<sup>137m</sup>.

Number of replicates ( <i>n</i> )	Rel. dead-time correction* $\left( \frac{F-1}{2F \ln F} R_0 \right)$ x 10 <sup>3</sup> τ	Mean value observed ( $\bar{t}_{\frac{1}{2}}$ ) min.	Precision of mean value ( $\sigma_{\text{rms}}[\bar{t}_{\frac{1}{2}}]$ ) min.
15	-38	2.573	0.002
15	-23	2.565	0.002
50	-15	2.564	0.001
30	-10	2.560	0.002
	( 0	2.557**	0.001 <sub>5</sub> )

\* confer equation (4)

\*\* by extrapolation

The two independent determinations of the half-life of Ba<sup>137m</sup> agree well (compare Tables 4 and 5) and the weighted grand mean is 2.5577 ± 0.0008 min. or 153.46 ± 0.05 sec., where the standard deviation given does not include radio-purital uncertainty. The systematic timing error, according

to expression (5), is less than or about equal to 0.0004 min., which may be disregarded.

The previously mentioned centre-interval,  $0.7 < t_{\frac{1}{2}}^{\ast\ast}/t_{\frac{1}{2}} < 1.7$ , corresponds to 1.8 min.  $< t_{\frac{1}{2}}^{\ast\ast} < 4.4$  min. in the case of  $\text{Ba}^{137\text{m}}$ . Since the stock solution of  $\text{Cs}^{137}$  had been aged for over a year before the half-life of  $\text{Ba}^{137\text{m}}$  was measured, an alien radio-nuclide belonging to this centre-interval could not have existed unless it sprang from a parent or forefather with a half-life of at least a month. An investigation of all known<sup>(16)</sup> radionuclides and their properties showed that only  $\text{Tl}^{208}$ ,  $\text{Tl}^{209}$  and  $\text{Bi}^{211}$  could have been of significance as centre-interval impurities. These three possibilities were investigated as follows.

$\text{Tl}^{208}$  and  $\text{Tl}^{209}$  emit photons of 2.6 and 1.6 MeV, respectively, whereas  $\text{Ba}^{137\text{m}}$  emits 0.7 MeV photons. In an analysis using a 1-channel  $\gamma$ -spectrometer no 2.6 or 1.6 MeV photons were detected in the radiation from  $\text{Ba}^{137\text{m}}$  samples. The limit of detection (i.e.,  $\sigma_{\Sigma^*}$ ) was less than 0.02 per cent of the total sum-count  $\Sigma$ , and the limit of the bias on  $t_{\frac{1}{2}}$  therefore less than 0.1 part in a thousand (cf. Table 2).

$\text{Bi}^{211}$  emits 5–6 times as many  $\alpha$ -particles as  $\gamma$ -photons, yet nuclear emulsions that were impregnated with drops of  $\text{Ba}^{137\text{m}}$  sample showed no  $\alpha$ -tracks above background after an exposure period equal to  $\theta$ . This investigation also established a limit for the possible amount of chain-contamination (cf. Table 3). By calibration with drops of uranium solution and by some conservative assumptions, the uncertainty of  $t_{\frac{1}{2}}$  due to the possibility of chain-contamination below the limit of detection was evaluated at 0.25 per mille, which is negligible compared to the above-mentioned  $\pm 0.0032$  min. due to the uncertainty involved in the investigation of wing-interval impurities.

The final result is thus  $2.5577 \pm 0.0032$  min., or

$$\underline{t_{\frac{1}{2}}[\text{Ba} - 137 \text{ m}] = 153.46 \pm 0.20 \text{ seconds.}}$$

## Silver 109m

### Previous Results

In 1941, HELMHOLZ<sup>(6)</sup> observed that the process  $\text{Ag}(d,2n)\text{Cd}$  lead to the production of a long-lived radio-isotope of cadmium (besides the already known 7-hour isotope), and a daughter of the long-lived isotope was found to have a half-life of  $40 \pm 3$  sec.

WIEDENBECK<sup>(7)</sup> reported in 1945 that silver nuclei excited by X-rays had a half-life of  $40.4 \pm 0.2$  sec. due to either  $\text{Ag}^{107\text{m}}$  or  $\text{Ag}^{109\text{m}}$ . The same

year, BRADT et al.<sup>(8)</sup> separated radio-silver from some aged radioactive cadmium, which had been produced by the process  $\text{Ag}(p,n)\text{Cd}$ , and found a  $t_{\frac{1}{2}}$  value of  $40.5 \pm 0.7$  sec., this was corrected the following year to  $39.2 \pm 0.2-0.3$  sec.<sup>(9)</sup>.

In 1947 BRADT et al.<sup>(10)</sup> announced that it had become possible, unequivocally, to attribute the  $t_{\frac{1}{2}}$  values 44 and 39 sec. to  $\text{Ag}^{107m}$  and  $\text{Ag}^{109m}$ , respectively, since HELMHOLZ<sup>(11)</sup> shortly before had made use of separated  $\text{Cd}^{106}$  and  $\text{Cd}^{108}$  that were irradiated with thermal neutrons.

Finally, in 1951, WOLICKI, WALDMAN and MILLER<sup>(12)</sup> excited a sample of highly purified  $\text{Ag}^{109}$  and measured the decay rate of the  $\text{Ag}^{109m}$  by photographing the sum-count and the clock every 5 sec. for 5 min. Their  $t_{\frac{1}{2}}$  value was graphically evaluated at  $40.0 \pm 1.0$  sec.

### Separation Technique

$\text{AgCl}$  was electroplated onto a disc of platinum gauze by the method of SUNDERMAN<sup>(13)</sup>, and  $\text{Ag}^{109m}$  in solution was separated from its parent,  $\text{Cd}^{109}$ , by exchange with some of the inactive silver ions in the net of  $\text{AgCl}$ .

### Present Result

74 half-life measurements were made (with  $F = 6$ ) using one of the previous counters reinforced by the linear amplifier of the  $\gamma$ -spectrometer in order to register the 22 keV X-rays from  $\text{Ag}^{109}$ . The dead-time of the set-up was about 10  $\mu\text{sec}$ . The main values that were found are given in Table 6.

TABLE 6. 74 measurements of the half-life of  $\text{Ag}^{109m}$ .

Number of replicates ( $n$ )	Total sum-count ( $\Sigma = R_0/\lambda$ ) kc.	Mean value observed ( $\bar{t}_{\frac{1}{2}}$ ) sec.	Precision of mean value ( $\sigma_{\text{rms}}[\bar{t}_{\frac{1}{2}}]$ ) sec.
25	150	40.08	0.04
49	25	39.85	0.05
	( 0	39.80*	0.06)

\* by extrapolation

The 49 measurements at  $\Sigma = 25$  kc. comprise 25 immediate measurements averaging  $39.89 \pm 0.07$  sec. and 24 delayed measurements (with

$2^N = 6$ ) averaging  $39.80 \pm 0.09$  sec. The difference between the immediate and the delayed series (i.e.,  $\Delta = 0.09$  sec.) is insignificant, since  $\sigma_{\text{rms}}[\Delta] = 0.12$  sec., and in consequence no wing-interval impurity was present in the samples. The uncertainty of this conclusion corresponds to  $\pm 1.5 \cdot \sigma_{\text{rms}}[\Delta] = \pm 0.18$  sec. on the  $t_{\frac{1}{2}}$  values (see page 8), and the final result is thus (cf. Table 6).

$$\underline{t_{\frac{1}{2}}[\text{Ag-109m}] = 39.80 \pm 0.18 \text{ seconds}}$$

pending the investigation of other possible impurities.

Since the  $\text{Cd}^{109}$  stock was aged for 9 months prior to the  $t_{\frac{1}{2}}$  measurements on the daughter, the only known<sup>(16)</sup> nuclide that could have been of significance as a centre-interval impurity was 30-sec.  $\text{Rh}^{106}$ , that emits 10% 0.5–3 MeV  $\gamma$ -photons. Making use of the 1-channel analyzer, the  $\text{Rh}^{106}$  content in the  $\text{Ag}^{109\text{m}}$  samples was found to be less than or about equal to 0.01 per cent which corresponds to a bias of less than 0.1 part in a thousand on  $t_{\frac{1}{2}}$  (cf. Table 2).

Finally,  $\text{Cd}^{109}$  is cyclotron produced, as opposed to  $\text{Cs}^{137}$  which is a fission product, so an investigation of chain-contamination was considered unnecessary in this case (cf. Table 3).

## Rhodium 106

### Previous Results

In 1946, SEELMANN-EGGEBERT<sup>(14)</sup> discovered a new radioisotope of ruthenium with a half-life of about 12 months. The emission of energetic  $\beta$ -particles suggested the existence of a short-lived daughter nuclide, and true enough a radio-isotope of rhodium with a half-life of about 40 sec. was found.

In 1951, GLENDENIN and STEINBERG<sup>(15)</sup> separated aged  $\text{Ru}^{106}$  from its daughter,  $\text{Rh}^{106}$ , by  $\text{HClO}_4$  distillation and found the half-life of  $\text{Rh}^{106}$  to be 25–30 sec. In one run they measured the decay of  $\text{Rh}^{106}$  remaining in the distillation flask ( $t_{\frac{1}{2}} = 30$  sec.), and in another run the in-growth of  $\text{Rh}^{106}$  in the distilled  $\text{Ru}^{106}$  ( $t_{\frac{1}{2}} = 31$  sec.). In a third run they filtered out the  $\beta$ -particles and the Bremsstrahlung from the  $\text{Rh}^{106}$  radiation and measured the  $\gamma$ -rays ( $t_{\frac{1}{2}} = 25$  sec.).

### Separation Techniques

Two different separation techniques were used. The one was Glendenin's distillation procedure modified so as to trap the distilled  $\text{RuO}_4$  in  $\text{HClO}_4$ ,

whereby it was ready for further distillation. The second technique was the absorption of  $\text{RuO}_4$  vapour as  $\text{RuO}_2$  on a paper disc held close to the surface of a saturated solution of  $\text{RuO}_4$  in 70%  $\text{HClO}_4$ .

### Present Result

The stock of  $\text{Ru}^{106}$  was aged for 8 months prior to the measurements on the half-life of  $\text{Rh}^{106}$ . In each measurement about 2 ml of Ru-distillation residue were rapidly taken, cooled and counted in a specially constructed flat-bulbed, V-shaped pipette provided with capillary stems. This container was stood on the aluminium window above a stilbene crystal for detection

TABLE 7. 23 measurements of the half-life of  $\text{Rh}^{106}$  by decay (given in chronological order).

Back-ground ( $B$ ) kc.	Rh-106 sum-count ( $\Sigma$ ) kc.	Average Rh-106 sum-count ( $\bar{\Sigma}$ ) kc.	Value observed ( $t_{\frac{1}{2}}$ ) sec.	Mean value and precision ( $\bar{t}_{\frac{1}{2}} \pm \sigma_{\text{rms}}[\bar{t}_{\frac{1}{2}}]$ ) sec.
25	87		30.9	
9	80		30.3	
22	66		30.8	
17	118		30.7	
11	46		30.1	
5	57		30.3	
8	46		30.3	
3	97		30.4	
5	77		30.6	
9	90		30.4	
7	51		30.5	
7	50		30.5	
4	46		30.5	
1	41		30.4	
6	64		30.4	
10	37		30.3	
3	48		30.9	
2	44		30.5	
5	42		30.8	
5	46		30.1	
3	51		30.7	
8	65		30.5	
2	55	61	30.8	$30.51 \pm 0.05$

of the  $\beta$ -particles from  $\text{Rh}^{106}$ . The special counting container circumvented a previously observed variation of the background counting rate due to translocation of undistilled  $\text{RuO}_4$  within the container during counting.

During 25 consecutive hours, 25 acceptable\* half-life measurements were made (with  $F = 4$ ) on the basis of 30 distillation cycles of a single portion of  $\text{Ru}^{106}$ . Two of the acceptable\* measurements were subsequently discarded, because the initial counting rate (calc. from  $\Sigma$ ) had exceeded 200 kc/min. and the linearity of the dead-time effect was therefore in doubt. The results of the remaining 23 measurements are given in Table 7.

A linear co-variance analysis of the values for  $t_{\frac{1}{2}}$  and  $\Sigma$  indicates a regression coefficient of  $0.0023 \pm 0.0024$  sec./kc. By extrapolation to  $\Sigma = 0$  (i.e.,  $R_0 = 0$ ) this conveys a dead-time correction on  $\bar{t}_{\frac{1}{2}}$  of  $-0.14 \pm 0.15$  sec. (cf. Table 7). Applying this correction, the resulting mean value for the half-life of  $\text{Rh}^{106}$  – as determined by decay – is  $30.37 \pm 0.16$  sec.

Each measurement of the half-life of  $\text{Rh}^{106}$  entailed a re-distillation of the stock of parent material, yet there is no chronological trend in the observed  $t_{\frac{1}{2}}$  values (cf. Table 7). In this case, radio-purital uncertainty is considered to have been insignificant in comparison with the above-mentioned uncertainty involved in the dead-time correction.

Finally,  $\text{Ru}^{106}$  was separated by the previously described paper-disc

TABLE 8. 10 measurements of the half-life of  $\text{Rh}^{106}$  by in-growth.

$R_1$	$R_2$	$R_\infty$	$F_{30}$	$\bar{F}_{30} \pm \sigma_{\text{rms}}[\bar{F}_{30}]$
6.3	9.4	12.6	1.97	
6.2	9.3	12.4	2.00	
4.5	7.3	10.1	2.00	
7.2	10.3	14.0	1.89	
4.7	7.0	9.4	1.96	
4.8	7.3	10.0	1.93	
5.4	8.4	11.2	2.06	
4.0	5.9	7.9	1.95	
5.1	7.6	9.8	2.14	
8.0	11.3	14.6	2.00	$1.990 \pm 0.022$

$R_1$  = number of kc. during the period from 0 to 15 sec.

$R_2$  = number of kc. during the period from 30 to 45 sec.

$R_\infty$  = number of kc./10 during the period from 450 to 600 sec.

$F_{30} = (R_\infty - R_1)/(R_\infty - R_2)$ .

\* The rather uncontrollable amount of undistilled  $\text{Ru}^{106}$  in the sample was sometimes unreasonably large. A measurement was deemed unacceptable in this respect (cf. p. 10) when the background sum-count exceeded one third of the  $\text{Rh}^{106}$  sum-count.



method from the solution of  $\text{RuO}_4$  which had been distilled 30 times, and 10 measurements on the rate of in-growth of  $\text{Rh}^{106}$  were made. In each measurement, the loaded paper disc was placed on the aluminium window of the detector and covered with an aluminium plate of the same thickness as the window. The data that were observed are given in Table 8.

The half-life of  $\text{Rh}^{106}$  – as determined by in-growth – may be calculated by use of the following equation (compare eq. (1)):

$$t_{\frac{1}{2}} = 30 \frac{\log 2}{\log F_{30}}.$$

Inserting the value of  $\bar{F}_{30}$  (cf. Table 8) we find the  $t_{\frac{1}{2}}$  value  $30.2 \pm 0.5$  sec.

The weighted mean of the value obtained by in-growth and the previously mentioned value obtained by decay gives the final result:

$$\underline{t_{\frac{1}{2}}[\text{Rh-106}] = 30.35 \pm 0.15 \text{ seconds.}}$$

### Acknowledgement

Financial support from the STATENS ALMINDELIGE VIDENSKABSFOND and KAI HANSENS FOND is gratefully acknowledged.

*Isotope Laboratory, Physics Department  
Royal Veterinary and Agricultural College,  
Copenhagen.*

---

## References

1. R. H. WASSERMAN, A. R. TWARDOCK and C. L. COMAR (1959), *Science* **129**, 568.
2. R. PEIERLS (1935), *Proc. Roy. Soc. A* **149**, 467.
3. A. C. G. MITCHELL and C. L. PEACOCK (1949), *Phys. Rev.* **75**, 197.
4. J. TOWNSEND, M. CLELAND and A. L. HUGHES (1948), *Phys. Rev.* **74**, 499.
5. D. W. ENGELKEMEIER (1948), ANL-4139 and AECD-2125.
6. A. C. HELMHOLZ (1941), *Phys. Rev.* **60**, 160.
7. M. L. WIEDENBECK (1945), *Phys. Rev.* **67**, 92.
8. H. BRADT, P. C. GUGELOT, O. HUBER, H. MEDICUS, P. PREISWERK und P. SCHERRER (1945), *Helv. Phys. Acta* **18**, 256.
9. H. BRADT, P. C. GUGELOT, O. HUBER, H. MEDICUS, P. PREISWERK, P. SCHERRER und R. STEFFAN (1946), *Helv. Phys. Acta* **19**, 218.
10. H. BRADT, P. C. GUGELOT, O. HUBER, H. MEDICUS, P. PREISWERK, P. SCHERRER und R. STEFFAN (1947), *Helv. Phys. Acta* **20**, 153.
11. A. C. HELMHOLZ (1946), *Phys. Rev.* **70**, 982.
12. E. J. WOLICKI, B. WALDMAN and W. C. MILLER (1951), *Phys. Rev.* **82**, 486.
13. D. N. SUNDERMAN and C. W. TOWNLEY (1961), "The Radiochemistry of Silver", USAEC NAS-NS **3047**, 25.
14. W. SEELMANN-EGGEBERT (1946), *Naturwiss.* **33**, 279.
15. L. E. GLENDENIN and E. P. STEINBERG (1951), *NNES-PRR* **9**, 793.
16. "Nuklidkarte", zweite Auflage (1961), Bundesminister für Atomenergie, Bad Godesberg.