Effect of Pressure on the Decay Constant of ^{99m}Tc

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An experimental investigation of the influence of high hydrostatic pressure on the decay constant of ^{99m}Tc has been performed. By the use of a multianvil high-pressure apparatus, a carrier-free source of ^{99m}Tc was compressed at a pressure of 100 kbar. The observed decay rate of the compressed ^{99m}Tc source was compared with that of a standard ^{99m}Tc source in the normal uncompressed state. The fractional increase in the decay constant, $\Delta\lambda/\lambda$, of the compressed ^{99m}Tc under that pressure was found to be $(4.6\pm2.3)\times10^{-4}$. Our result agrees with that observed by Bainbridge, $\Delta\lambda/\lambda=(2.3\pm0.5)\times10^{-4}$, to within a factor of about 2. Comparison with theoretical studies by other workers is also discussed.

I. INTRODUCTION

The change in a nuclear decay constant, λ, by any external effect has been observed by many workers since the interesting suggestions by Segrè[1] and Daudel[2] in 1947. As is well understood, the decay types which may give rise to observable changes in λ are those in which the orbital electrons involved contribute directly to the nuclear decay, i.e., either by an orbital-electron capture or by a partly converted isomeric transition of a nucleus. Small changes in λ values have been found to result from the effects of different chemical states of combinations on particular radioactive substances.[3-10]

The first attempt to find a high-pressure effect on a nuclear decay rate was by Bainbridge[11] using ^{99m}Tc under a pressure of 100 kbar. He found that the relative change in the decay constant of ^{99m}Tc, $\Delta\lambda/\lambda$, was $(2.3\pm0.5)\times10^{-4}$. Unfortunately, the details of the work have never been published. Gogarty, Kistler, and Christiansen[12] studied ^{7}Be and ^{131}Ba, where both samples were subjected to pressures of 157 kbar for 20 days and 140 kbar for 48 days. From their results, it was revealed that the expected values of $\Delta\lambda/\lambda$ at a pressure of 100 kbar are 2×10^{-3} for ^{7}Be and 0.66×10^{-3} for ^{131}Ba. Cooper[13] measured $\Delta\lambda/\lambda$ of ^{90m}Nb also under a pressure of 100 kbar and obtained the value of $(6.3\pm7)\times10^{-3}$.

It seems worthwhile to provide additional information on the high-pressure effect on a nuclear decay rate. Although there are several possible nuclides which could be used in the study, the selection of these depends on the characteristics of the high-pressure device, for example, time required to generate a high hydrostatic pressure and to achieve geometrical stability of the device.

A multianvil (eight-anvil) high-pressure apparatus, developed by Kawai[14] and modified in some details was used in the present experiment. The main modification of the apparatus made was to improve the geometrical stability and the solid angle for γ-ray detection. ^{99m}Tc atoms were compressed under a pressure of 100 kbar with this apparatus. Since the transition of ^{99m}Tc is a 2.17-keV $E3$ isomeric transition ($T_{1/2}=6.04$ h), the predominant process of the decay is internal conversion. The contributions to the conversion process come from M and outer shells. This indicates that the external effect, pressure effect or otherwise, on the decay rate of the nuclide might be large enough to be measurable.

Since the decay rate of the level is measured by detecting the 140-keV γ rays immediately following the isomeric transition involved, there must be a slit in the high-pressure apparatus, through which the γ rays can come out. By comparing two carrier-free ^{99m}Tc sources, one under a pressure of 100 kbar and the other in the uncompressed state, the relative change in their intensities was observed as a function of time.

In the present paper, we wish to report details of our experimental work with ^{99m}Tc, as well as some characteristics of the high-pressure device adopted in this work.

II. PRINCIPLE OF THE MEASUREMENT

In general, a single measurement of a decay curve is not a satisfactory way to determine any change in λ caused by different electronic environments. The so-called *differential method* pioneered by Rutherford[15] is the most appropriate one for determination of $\Delta\lambda/\lambda$. The attractive features of this method are that two sources are measured at the same time and the systematic errors of the measuring system can be minimized by exchanging the sources. In the high-pressure experiment, however, one cannot exchange the

compressed source with the standard uncompressed one. In order to minimize the systematic errors, we prefer to use only one detector instead of two as in the differential method, and to measure alternately two sources which give almost the same counting rate. Then the difference of two measured counts accumulated in a certain time interval is assigned as the output at time t, where we assume t to be the same for both samples.

Supposing that the decay constant of the standard ^{99m}Tc source is λ and the constant of the compressed source at 100 kbar is $\lambda+\Delta\lambda$, the difference of the counting rates of the two sources is given as follows:

$$N(t)=N_s(t)-N_c(t)$$
$$=[N_s(0)-N_c(0)]e^{-\lambda t}+N_c(0)\Delta\lambda te^{-\lambda t}, \quad (1)$$

where the subscripts s and c refer to the standard and compressed sources, respectively. By multiplying Eq. (1) by $e^{\lambda t}$, one gets

$$N(t)e^{\lambda t}=[N_s(0)-N_c(0)]+N_c(0)\Delta\lambda t. \quad (2)$$

Since Eq. (2) has a linear form $y=a+bt$, the slope of a plot of $N(t)e^{\lambda t}$ versus t is equal to $N_c(0)\Delta\lambda$, from which $\Delta\lambda/\lambda$ can be evaluated.

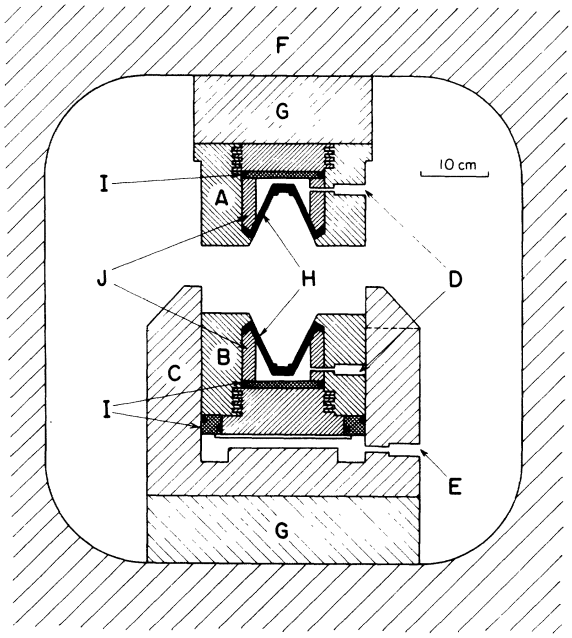

FIG. 1. The construction of the multianvil high-pressure devide: (A) and (B) inner cylinders; (C) outer cylinder; (D) oil inlets of inner cylinders; (E) oil inlet of outer cylinder; (F) iron frame; (G) iron bases for cylinders; (H) rubber caps which shield the cylinder oil as well as hold the anvils (not shown in the figure); (I) oil shields made of nylon gaskets and rubber O rings; (J) iron cylinders to hold the rubber caps.

In order to insure the stability of the whole measuring system, we used a third source, ^{57}Co, and measured the 122-keV γ rays from the second excited level of the daughter nucleus, ^{57}Fe. As the source is expected to keep a constant intensity during measurements, the counts of the 122-keV γ rays are used as a correction factor for $N_s(t)$ and $N_c(t)$.

Since the ^{99m}Tc source to be compressed is embedded in a pressure-transmitting medium [pyrophyllite, $Al_2Si_4O_{10}(OH)_2$], there might be some chemical or structural effect on λ when the sample is compressed under a high pressure. To distinguish a pressure effect, not accompanied by the other effect, the generated hydrostatic pressure was decreased to 1 atm after the measurements of $N_s(t)$ and $N_c(t)$ were completed. Then the decay probabilities of the standard and decompressed ^{99m}Tc sources were compared by the procedure mentioned above.

In this work a digital method was adopted instead of an analog method which we would have preferred to have used. The differential analog method can be used conveniently with strong sources. We would prefer to record the values of $N(t)$ by analog values for the following three reasons: (a) Balancing of two outputs can be achieved by using a differential amplifier, (b) the gain of the differential amplifier can be large, and (c) no dead-time correction is required. But in the present situation the measurements had to be made alternately, the counting rates are not extremely large, and there is therefore no reason to convert a digital to an analog value. The only additional error introduced in a digital method is due to a dead-time correction, which will be discussed in Sec. IV D.

III. HIGH-PRESSURE DEVICE

A. Construction

The multianvil high-pressure device used consists of eight anvils, three cylinders, and an iron frame in which the cylinders are contained. In Fig. 1 are shown the details of the construction, where the anvils are embraced in the rubber caps denoted as H in the figure.

Each of the anvils has two components, a hardened iron section and a well-sintered tungsten carbide section containing a few percent cobalt (Fig. 2). Four anvils then compose a cone with four appropriate side boards and a cap, both made of iron (Fig. 3). Between each anvil are sandwiched several small spacers of 1.2-mm thickness which were prepared by mixing powder of pyrophyllite and Araldite in the weight ratio 1:1. Since the top of tungsten carbide section of each anvil is truncated so as to have a front face of an equilateral triangle having sides of 1.8 mm, an

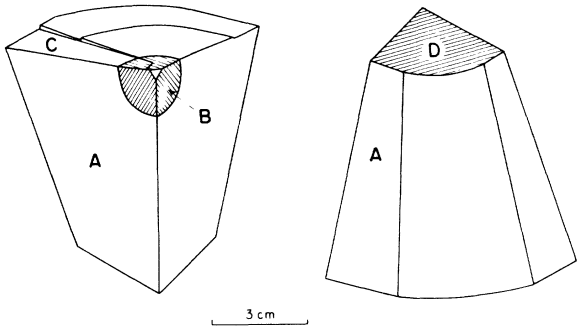

FIG. 2. Anvil: (A) hardened iron section; (B) tungsten carbide section with truncated top; (C) small opening to obtain a larger γ-ray detection efficiency; (D) lead plate of 0.5-mm thickness.

octahedral hollow space is formed at the center when all anvils are put together. In the hollow space is inserted an octahedron of pyrophyllite with edges of 3.7 mm. In order to embed a point source, the octahedron is formed from two pieces.

The cones are then mounted in thick rubber caps which shield the cylinder oil as well as hold the cones at the proper position in the assembly. When the cylinder oil pressure is elevated, the cones are squeezed toward the center until the force causing the outward flow of the central pyrophyllite is balanced with the internal shearing stress of the assembly.

As illustrated in Fig. 1, cylinder A is fixed on the iron frame which can withstand a maximum load of 900 tons. Cylinder B can move upward and downward in cylinder C which is also fixed on the frame. There are two independent oil systems, one connected in parallel to the inner cylinders, A and B, and the other to cylinder C. These systems are fitted with pressure-recording units to control and monitor oil pressures. After two cones are mounted in the inner cylinders, cylinder B is elevated until the space between the two inner cylinders becomes 5 mm. Then two pumps (maximum available pressure, 700 kg/cm^2) are operated alternately until a pressure of 100 kbar is generated at the center of the octahedron. When the anvils move toward the center by elevating the oil pressure, the octahedral pyrophyllite at the center is compressed and part of it becomes a gasket between the neighboring anvils preventing outflow of the inside material. At the final stage of compression, the slit between two inner cylinders is decreased to about 3.5 mm.

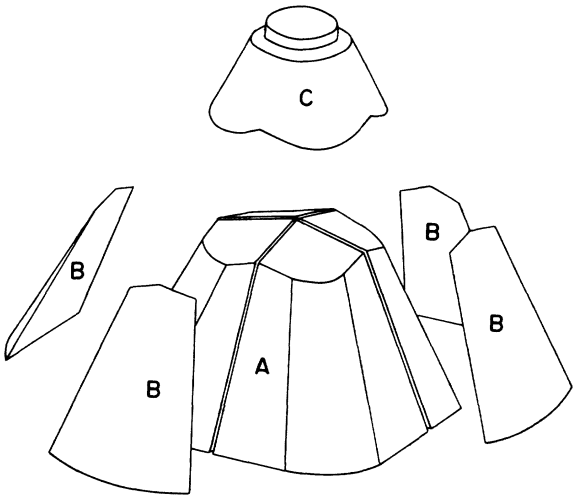

FIG. 3. Cone which consists of four anvils (A), four side boards (B), and a cap (C).

B. Pressure Calibration

Pressure calibration of the present device is based upon the phase transitions of bismuth and tin. The phase transitions of Bi I-II, Bi III-V, and Sn I-II are known to correspond to pressures of 25.2, 77.0, and 100.0 kbar, respectively.

Prior to the experiment, we measured the relation between the oil pressure in cylinders A and B, and the generated hydrostatic pressure. A thread of bismuth or tin is sandwiched between two pieces of pyrophyllite which together form an octahedron. Both ends of the thread are terminated to thin copper films which are individually connected through wires to an apparatus for measuring electric resistance. Changes in resistance of the bismuth or tin thread caused by the phase transitions are recorded.

As shown in Fig. 4, repeated measurements give almost a linear relation between the oil and the hydrostatic pressures. From the result obtained, it was found that a pressure of 100 kbar corresponds to 520 ± 50 kg/cm^2. The reproduci-

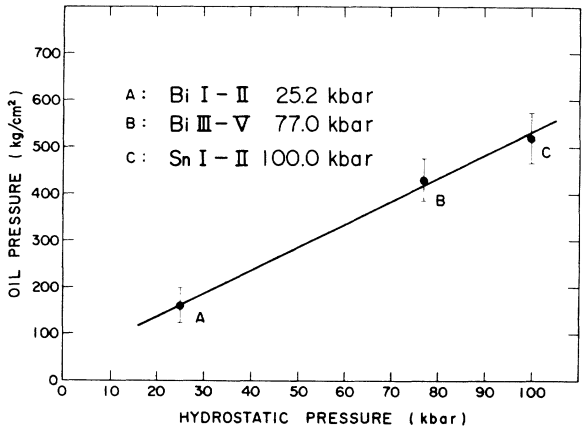

FIG. 4. Oil pressure in the inner cylinders versus generated hydrostatic pressure.

bility of measurements is rather poor, but this seems to be unavoidable since in each measurement the anvils are put in place by hand. The following experiment was performed under an oil pressure of 520 kg/cm^2.

C. Stability

The most important and critical feature of the present experiment is the geometrical stability of the pressure device. The generated hydrostatic pressure itself is certainly quite stable because of the large hysteresis of pressure yield. However, as the γ rays orginating at the center of the pressure device are measured outside the cylinders, the observed counting rate of the γ rays is crucially affected by the geometrical detection efficiency. If there is any slight change in the geometry of the assembly during measurements, the experiment would be meaningless.

The geometrical stability was carefully examined using both ^{99m}Tc and ^{57}Co sources embedded in the octahedral pyrophyllite. Figure 5 shows the counting rates of the 140-keV γ rays from the ^{99m}Tc source compressed at 100 kbar and measured by a NaI(Tl) scintillation detector set outside the pressure device. For comparison the counting rates of the γ rays from the standard ^{99m}Tc source are also shown. The data are corrected for decay and dead time of the counting system. It was found that several hours are required before the necessary geometrical stability is achieved. Considering these preliminary examinations, experiments for observing the fractional change in the decay constant of ^{99m}Tc were started at least six hours after the sample was compressed at a pressure of 100 kbar.

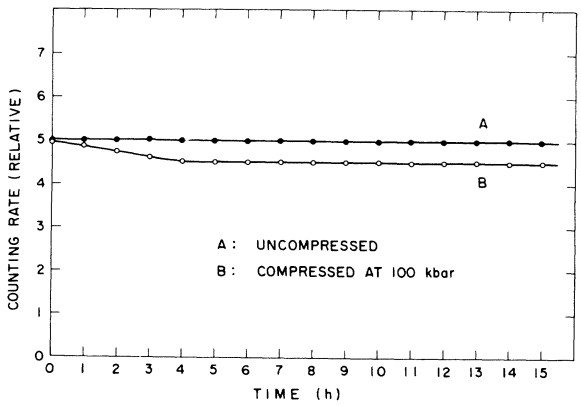

FIG. 5. Geometrical stability of the high-pressure device (^{99m}Tc source). Data are corrected for decay and dead time of the counting system. For comparison, observed counting rates of the standard-uncompressed ^{99m}Tc source are also shown.

IV. EXPERIMENTAL PROCEDURE

A. Source Preparation

The ^{99m}Tc solution was obtained by milking a commercial generator containing the longer-lived parent ^{99}Mo. The initial intensity of ^{99}Mo was 100 mCi. Only technetium as pertechnetate ions is eluted when a solution of NaCl is passed through the column loaded with ^{99}Mo. The residue remaining on evaporation of the eluted solution is expected to be rather bulky mainly owing to NaCl. Since the ^{99m}Tc source to be embedded in the central octahedron should not be bulky, it was necessary to prepare the salt-free solution before the final source preparation. For this reason, the following chemical procedures are carried out: (a) Dissolve 6 g of KOH in 20 ml of solution eluted from the ^{99}Mo column and transfer the solution into a 50-ml separation funnel; (b) add 4 ml of pyridine to the funnel and shake for 30 sec; (c) after the phase separation is completed, separate the organic phase; (d) to remove trace cations, such as Na^+ and K^+, pass the organic phase through a cation-exchange column packed with Dowex 50W X8 (100–200 mesh, 1.5 cm diam by 5 cm long) and preliminarily conditioned with 5% NaOH, 5% HCl solutions, and distilled water; and (e) evaporate the solution eluted from the cation-exchange column almost to dryness.

With the condensed radioactive solution of ^{99m}Tc thus obtained, a point source of ^{99m}Tc was prepared at the center of the octahedral pyrophyllite. As mentioned before, the octahedron was divided into two pieces, each of which had a small hole of 1-mm diameter at the center of the larger face. After the residue from evaporation was mounted in these central holes, the pyrophyllite pieces were inserted in a quartz tube filled with flowing oxygen-free nitrogen or hydrogen, one section of which was heated in an electric furnace. The oxygen-free nitrogen was prepared by passing it through an activated copper column. In this quartz tube, the source was reduced in a flowing hydrogen atmosphere at 600°C for 1 h, to ensure oxygen-free technetium. It is quite certain that by this heating procedure zeolitic water molecules contained in the backing pyrophyllite can be removed. When the reduction procedure was completed, the pyrophyllite pieces were attached in a nitrogen atmosphere to form an octahedron as well as to prevent tarnishing of the technetium in air, and then transferred into the high-pressure device. The source thus prepared had an initial intensity of about 10 mCi.

The standard source of ^{99m}Tc was prepared at the same time by the procedure described above. A disk made of pyrophyllite, 1 cm diam by 1 mm

thick and having a small cavity of 1-mm diameter, was used as the backing material in order to secure the same chemical form of ^{99m}Tc with the point source to be compressed. After reduction in a hydrogen atmosphere at 600°C for 1 h, the disk source was tightly wrapped with vinyl tape in a nitrogen atmosphere at room temperature, so that the technetium was expected to keep the same chemical form during measurements. The disk source was then mounted on a slide bench with a micrometer screw by which the position of the standard source could be precisely controlled. The intensity of the source was chosen to be about $\frac{1}{10}$ of the compressed source because of the different geometrical detection efficiencies. By varying the aperture of the standard disk source from the detector, the initial counting rates of both sources, compressed and standard, were balanced taking into account the time lag of measurements in the experiment, so that the difference of the initial counting rates of both sources, $N_s(0) - N_c(0)$, was kept almost as low as several times the statistical error involved.

Another source of ^{57}Co used for checking the stability of the whole measuring system had an intensity of about 1 mCi. The source was inserted in the face of a lead brick that served as a source holder and shield. No special treatment was necessary for this source.

B. Detection System

The detector used for the 140- and the 122-keV γ rays is an 1.5-in.-diam by 1-in.-thick NaI(Tl) scintillator. Of particular importance in the present work is the stability of the photomultiplier in view of the long times required to accumulate counts. To obtain maximum stability, careful examinations of 10 Toshiba 7696 phototubes were carried out. The most stable phototube adopted had a long-term instability of less than ±0.4%/3 days operated at −700 V. The pulses from the photomultiplier were fed to a single-channel pulse-height analyzer through a preamplifier and a main amplifier, and then accumulated in a 5-MHz scaler.

The whole electronic system described above was used alternately for measurements of the γ rays emitted from the three kinds of sources, i.e., the compressed or decompressed ^{99m}Tc, the standard ^{99m}Tc, and the ^{57}Co. The probe in which the NaI(Tl) scintillator, the photomultiplier, and the preamplifier were housed was set on a turntable in order to perform individual measurements of each source with satisfactory reproducibility in positions. This setup allowed one to perform alternate measurements of the three sources without moving them. Each source was properly shielded so as to prevent the counter from seeing the other sources. Furthermore, the probe of the counter was covered by a lead shield 6 mm thick. Thus, we were able to carry out completely independent measurements of the source in question without any excess counts due to the other sources.

C. Measurements of $N(t)$

Six hours or more after compression of the point source embedded in the octahedral pyrophyllite, the number of the 140-keV γ rays detected by the NaI(Tl) scintillator was accumulated in a 5-MHz scaler. The initial counting rate was chosen to be about 20 000 cps. To achieve good statistics, each measured value of $N_c(t)$ in Eq. (1) was given by the average value of the total counts for the time period $t \pm 12$ min. Then the counter was turned toward the ^{57}Co source and 1 min after the end of the preceding measurement, the 122-keV γ rays from the source were accumulated for 9 min. By turning the counter toward the ^{99m}Tc standard source, we found $N_s(t)$, starting 1 min after the ^{57}Co measurement was completed and continuing for 24 min as in the measurement of $N_c(t)$. These three measurements supplied a set of data and this entire counting procedure was repeated nine times. The total time for each run was 10 h and runs were repeated 23 times.

After each run was completed, the pressure was decreased to atmospheric pressure and comparisons of the standard and decompressed ^{99m}Tc sources were made by the same procedure described above. When the pressure was decreased, the geometrical detection efficiency for γ rays was increased but only by a factor of about 1.1. To obtain similar initial counting rate to that of the preceding measurement, the slit between two inner cylinders had to be slightly enlarged. Then the position of the standard ^{99m}Tc source was again adjusted to balance both counting rates.

D. Dead-Time Correction

The most troublesome source of errors is uncertainty in the resolving time, τ, of the whole measuring system. Without knowing the precise value of τ, the digital method cannot be applied.

If two ^{99m}Tc sources to be compared could be balanced with sufficient accuracy, the difference of their initial counting rates, $N_s(0) - N_c(0)$ in Eq. (2), becomes nearly zero and consequently, uncertainty in τ does not cause a significant error in the evaluation of $\Delta\lambda/\lambda$. However, balancing could be achieved at best only to within several times the statistical error of $N_s(0)$ or $N_c(0)$, even by adjusting carefully the position of the standard source mounted on the slide bench with a microm-

eter screw. Supposing that $\Delta\lambda/\lambda$ is 1×10^{-4} and $N_c(0)$ is 20 000 cps, the last term in Eq. (2), $N_c(0)\Delta\lambda t$, gives about 2 cps at $t=10$ h. Considering the expected value of $N_c(0)\Delta\lambda t$ and the actual nonzero value of $N_s(0)-N_c(0)$, the upper limit of uncertainty in τ was estimated to be a few percent.

In order to determine τ with an error of a few percent at maximum, the decay curves of ^{99m}Tc multiplied by $e^{\lambda t}$ were measured repeatedly by the present measuring system under normal conditions. Without dead-time corrections, curves showed positive slopes due to more counting loss at higher counting rate. Then by applying a least-squares fit and the standard equation for dead-time correction, τ was evaluated so as to make the curves horizontal straight lines. The most probable value of τ thus obtained and its standard deviation were determined as $\tau=(2.37\pm0.05)\times10^{-6}$ sec. The dead-time correction for all data was done with this value of τ.

V. RESULTS AND DISCUSSION

As indicated by Eq. (2), the observed value of $N(t)$ multiplied by $e^{\lambda t}$ and corrected for the resolving time gives a linear form. From the slope of a least-squares curve of $N(t)e^{\lambda t}$, one can get the value of $\Delta\lambda/\lambda$. As a standard of comparison, the decay constant of ^{99m}Tc, $\lambda=0.1148\pm0.0005$ h^{-1}, measured by Bainbridge, Goldhaber, and Wilson[4] was used. The individual experimental results of $\Delta\lambda/\lambda$ obtained for pairs of standard-compressed ^{99m}Tc sources had standard deviations of about 100%. Nevertheless, they were found mostly to have positive values of the order of 10^{-4}. On the contrary, the $\Delta\lambda/\lambda$ values obtained for combinations of the standard and decompressed ^{99m}Tc

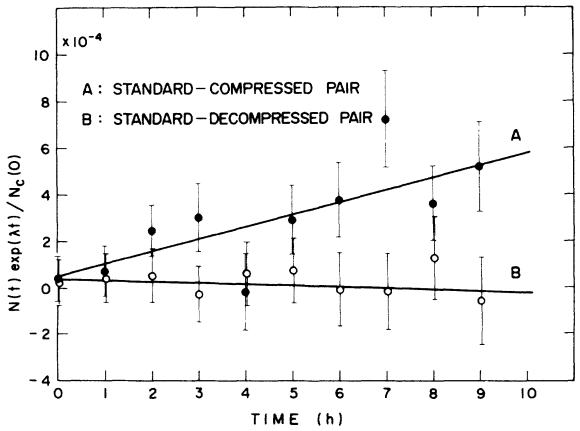

FIG. 6. Differences of counting rates of the standard-compressed pair of ^{99m}Tc sources and the standard-decompressed pair. Data are multiplied by $e^{\lambda t}$ and divided by the initial counting rate of the compressed or decompressed source, respectively.

sources were distributed around zero. This fact leads us to conclude that there is a minute but apparent increase in λ of ^{99m}Tc caused by the effect of high pressure.

There are some factors that could cause errors in the determination of $\Delta\lambda/\lambda$. The magnitudes of errors are given as fractions of $N_c(0)$: (a) statistical fluctuations in $N(t)=N_s(t)-N_c(t)$, less than 0.02% at $t=0$; (b) errors introduced by the exponential decrease of source intensity when $N_s(t)$ or $N_c(t)$ is obtained as an average value for time duration $t\pm12$ min, about 0.01%; (c) error in time measurements, negligible; (d) errors due to gain drift of the whole measuring system and wall contamination, which were determined experimentally to be 0.05%; (e) uncertainty in λ; and (f) uncertainty in the generated hydrostatic pressure which is discussed in Sec. III B.

Considering these errors and summarizing all data, the final experimental results are obtained as shown in Fig. 6, where least-squares curves of $N(t)e^{\lambda t}/N_c(0)$ for the standard-compressed pair and the standard-decompressed pair are indicated. The values of $\Delta\lambda/\lambda$ obtained from the results are $(4.6\pm2.3)\times10^{-4}$ for the standard-compressed pair and $(-0.2\pm1.1)\times10^{-4}$ for the standard-decompressed pair. From the results, we believe that within the experimental error there is no chemical or structural effect which might by caused by compression and that the backing pyrophyllite without zeolitic water molecules is in the present sense not active chemically at 100 kbar and at room temperature. It seems to be plausible to conclude that the larger value for the standard-compressed pair can be attributed to the relative increase of the decay constant of ^{99m}Tc under a pressure of 100 kbar. The pressure effect presented here is large compared with Bainbridge's,[11] $(2.3\pm0.5)\times10^{-4}$, by a factor of about 2. But, because of the rather large experimental error of the present result, it may be said that our result agrees with that observed by Bainbridge.

In order to pursue theoretical estimates of the pressure effect on λ, one has to find the relative contributions of the different electron levels to the internal-conversion coefficient, α, and consequently to the decay rate λ. It was found by Porter and McMillan[16] that the main contribution to α comes from the $3p$ and $3d$ levels, while the principal contribution to the change in α by a hydrostatic pressure comes from the $4p$, $4d$, and $5s$ levels. By the use of the Thomas-Fermi statistical potential applied to metallic ^{99m}Tc and corrected for the electron self-potential, they obtained the wave functions of the different electron levels involved and the energy term values were determined for metallic ^{99m}Tc in the normal un-

compressed state and in the 10% compressed state. Assuming the internal-conversion coefficient to be linear in pressure and the compressibility to be 0.27 $Mbar^{-1}$, they concluded the fractional change in λ of metallic ^{99m}Tc to be $(2 \text{ to } 4)\times 10^{-4}$ under a pressure of 100 kbar.

It should be mentioned here that the relative contributions of different electron states to λ of a free ^{99m}Tc atom have also been calculated by Slater,[17] where he used $1/r^4$ for the radial dependence of electromagnetic field instead of the spherical Hankel function as used by Porter and McMillan.[16] The relative contributions of the M and N shells to λ are roughly 87 and 13% by Slater and 91 and 6% by Porter and McMillan, respectively. These results obtained independently seem to diverge appreciably from each other, even taking into consideration the different assumptions.

The M and N subshell ratios of the 2.17-keV transition in ^{99m}Tc were observed by Amtey, Hamilton, and Zender,[18] and recently in the same laboratory more refined measurements were performed by Lacasse and Hamilton.[19] These experiments were carried out with mass-free ^{99m}Tc sources which may be approximated by a free-atom model. They found that the contribution of M-shell electrons to λ is about 88%, while that of the N-shell electrons is about 12%. This result agrees fairly well with that obtained by Slater.[17]

The experiment presented here was done with carrier-free ^{99m}Tc sources. Referring to these four investigations by other workers, we feel that, for comparison with the present result, a refined theoretical estimation of the pressure effect on λ using the free-atom model is needed. Theoretical work in this direction is being pursued.

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