Precision measurements of the half-life of ^{196}Au and ^{202}Tl

By K. Hirose^{1,*}, H. Kikunaga² and T. Ohtsuki¹

¹ Research Center for Electron Photon Science, Tohoku University, Sendai 982-0826, Japan

² Graduate School of Science, Osaka University, Toyonaka, Osaka 560-0043, Japan

(Received December 21, 2009; accepted in final form October 19, 2010)

Half-life / Radionuclides Au-196, Tl-202

Summary. The half-lives of ^{196}Au and ^{202}Tl have been precisely obtained by repeated measurements for 40 samples of ^{196}Au and 16 samples of ^{202}Tl using germanium detectors. The dead-time was corrected by the simultaneous detection of 662-keV γ rays from a reference source of ^{137}Cs . The half-life value of 6.1451 ± 0.0013 (tot.) d, 0.35% shorter than the recommended value of ENSDF [1], has been obtained for ^{196}Au . The error contains the statistical uncertainty and other random uncertainty. The half-life of ^{202}Tl is determined to be 12.4706 ± 0.0055 (tot.) d with improved accuracy as compared to the latest measurement by a factor of 15.

1. Introduction

Half-lives of many radioactive isotopes have been measured with relatively small uncertainties (for example [2, 3]). There are some measurements with an uncertainty of the order of 0.01% [3]. Since it contains, in many cases, only statistical error, other random uncertainties and systematic uncertainties are important. In the present study, the half-lives of ^{196}Au and ^{202}Tl were precisely measured using a high-purity germanium (HPGe) detector coupled with an automated sample changer (ASC) which has been also used in the investigation of the half-life change of ^7Be [4].

It has been reported that the changes in the half-life of $^{99\text{m}}\text{Tc}$ due to its chemical forms, temperature and pressure were in a range of 10^{-2} – $10^{-1}\%$ [5–11]. The magnitude of the half-life change is expected to be very small. Therefore, the evaluation of the uncertainty is important for such studies. In the present measurement, 40 and 16 samples of ^{196}Au and ^{202}Tl were measured in order to reduce and estimate the random uncertainty. From the estimated random uncertainty, we evaluate whether the system can be used for the investigation of the potential changes in half-lives.

2. Experiment

The ^{196}Au and ^{202}Tl samples were produced by the (γ, n) reaction at the Research Center for Electron Photon Science, Tohoku University. These nuclides were selected because there are only a few measurements with high accuracy. Furthermore, their half-lives are adequately long enough to ac-

cumulate sufficient statistics without large dead-time. Gold foils (typically 20 mg) and Tl_2O_3 samples (150 mg) were irradiated by the bremsstrahlung photons with an end-point energy of 30 MeV. These samples were irradiated for about 10 h at an electron beam current of about 130 μA . The irradiated Tl_2O_3 powder was put into an aluminum case of 7 mm in diameter and 6 mm in depth, and fastened with epoxy adhesive in order to fix the counting geometry.

The γ rays from the samples were detected using a HPGe detector coupled with ASC. In the present study, four pairs of the HPGe detector and ASC were used in order to estimate and reduce random uncertainties. Two samples (16 pairs of ^{196}Au – ^{202}Tl or 12 pairs of ^{196}Au – ^{196}Au) mounted on the arms of ASC were alternately located in front of the HPGe detector according to a time period. The distances between the sample and the end-cap of the HPGe were typically 30 mm. Since each ASC was driven by a servomotor with a positional accuracy of 0.02 mm, the counting geometry was not affected by the sample changing. For the dead-time correction, a reference source of ^{137}Cs , purchased from Japan Radioisotope Association, was placed in a fixed counting geometry. The energy spectra for each sample were measured for one or two hours (3550 or 7150 seconds for accumulation of a γ -ray spectrum and the remaining 50 seconds were reserved for the sample changing). The dead-time of the data-acquisition (DAQ) system was typically 10% at the beginning and became a few % at the end of the measurements. The time clock of the DAQ system was periodically calibrated using the Japan standard time distributed *via* a long-wave radio signal. The decays of 40 samples of ^{196}Au and 16 samples of ^{202}Tl were followed for several half-lives.

3. Results and discussion

A typical energy spectrum around 356 keV of ^{196}Au is shown in Fig. 1. The dashed curve is a fitting result by a combination of a gaussian and error function. A peak region was determined as $\pm 3\sigma$ around the center of the peak. The background was estimated from the counts in 2σ -width regions on both sides of the peak.

Then, the peak area $P(t)$ at time t is determined as,

$$P(t) = \sum_{\text{Peak}} C(t) - \frac{3}{2} \sum_{\text{BG}} C(t). \quad (1)$$

* Author for correspondence (E-mail: hirose@ins.tohoku.ac.jp).

Fig. 1. The pulse height spectrum around 356 keV for ^{196}Au . The dashed curve is a fitting result by a gaussian and error function. The peak area is counted in $\pm 3\sigma$ around the peak center (hatched with dark gray). The backgrounds are estimated from the 2σ -width regions on both sides of the peak (hatched with light gray).

The peak areas of the 333 (^{196}Au), the 440 (^{202}Tl) and the 662-keV (^{137}Cs reference source) γ rays were determined in the same manner.

In the present work, a reference method using a ^{137}Cs source was employed for dead-time and pile-up corrections. In this method, the half-life is obtained from the ratio of the activity of interest ($Ae^{-\lambda t}$) to that of the reference source ($A'e^{-\lambda't}$). However, the ratio of the counts integrated over the finite time period is obtained in practice. Therefore, the ratio is given as,

$$\frac{P(t)}{P'(t)} \equiv R(t) = \frac{\int_t^{t+\Delta t} Ae^{-\lambda x} \eta(x) dx}{\int_t^{t+\Delta t} A'e^{-\lambda' x} \eta(x) dx}, \quad (2)$$

where Δt is a measurement period for each data point (3550 or 7150 s in this work) and η is the ratio of live time to real time. If the activity of the reference source is constant in the time interval Δt , $\eta(t)$ is expressed as $a(1 - be^{-\lambda t})$. This is valid considering that the dead-time is proportional to the rate of input signals. The initial value of η is $a(1 - b)$ and a is smaller than unity because of the activity of the reference source. Then, one can calculate $R(t)$ as,

$$R(t) = \frac{Ae^{-\lambda t}}{A'e^{-\lambda' t}} \cdot \frac{1 - e^{-\lambda \Delta t} - \frac{b}{2}(1 - e^{-2\lambda \Delta t})e^{-\lambda t}}{\lambda \Delta t - b(1 - e^{-\lambda \Delta t})e^{-\lambda t}} \quad (3)$$

The last factor of this equation is a monotonically increasing function with time t . However, the time dependence of this factor is extremely small. For example, assuming $\lambda = 4.8135 \times 10^{-3} \text{ h}^{-1}$ (6 d), $\Delta t = 2 \text{ h}$ and $b = 0.5$ (corresponds to 50% initial dead-time), the difference of this factor is only $0.8 \times 10^{-3}\%$ between at $t = 0$ and at $t = \infty$. Therefore, we can put this factor constant and derive the half-life from $R(t) \propto Ae^{-\lambda t} / A'e^{-\lambda' t}$.

A typical decay curve obtained from the 356-keV γ ray of ^{196}Au is shown in the upper panel of Fig. 2. The thin line is the result of the least-square fitting by an exponential function. The lower panel shows the deviations from the fitted function, divided by the statistical errors. The reduced chi-

Fig. 2. The upper panel shows the decay curve for the 356 keV γ ray of ^{196}Au . The thin line is the fitting result by an exponential function. The lower panel shows the normalized residuals.

Fig. 3. The half-life values of (a) ^{196}Au and (b) ^{202}Tl . The center panels show the all results obtained in the present work (closed and open circles in the figure (a) are deduced from the 333 and 356-keV γ rays, respectively). The half-lives are determined by the least-square method. The deviations are shown in the right panels. In the left panels, our results (closed circles) are compared with the other data referred from [2, 3, 12–21].

squares for 40 runs were around unity (0.83 at minimum, 1.67 at maximum), which shows the validity of the data and statistical errors being fitted by an exponential function.

The half-lives of ^{196}Au and ^{202}Tl are shown in Fig. 3a and b, respectively. The center panels show the all results obtained in this work. The results from the 333 and the 356-keV γ rays of ^{196}Au are plotted by closed and open circles, respectively. The values of the half-lives were determined by the least-square fitting with a constant parameter,

i.e., 0-order monomial. For the half-life of ^{196}Au , 6.1512 ± 0.0003 (sta.) d and 6.1459 ± 0.0002 (sta.) d were obtained from the 333 and the 356-keV γ rays, respectively. The difference between them is much larger than the statistical uncertainty. The half-life of ^{202}Tl was determined to be 12.4728 ± 0.0006 (sta.) d.

The reduced chi-squares in these fittings, however, were 47.6 and 48.3 for ^{196}Au and 75.3 for ^{202}Tl , which means that the half-lives obtained from the repeated measurements contained not only the statistical error but also other random errors. As can be seen from the right panels of Fig. 3, the deviations from the half-lives are much larger than the statistical error of each data point, although there are some data points with large statistical errors. From these deviations, the root mean squares were found to be 0.015 d (0.25%) and 0.0074 d (0.12%) for the 333 and the 356-keV γ rays of ^{196}Au , respectively, and 0.021 d (0.17%) for ^{202}Tl . Hence, the magnitude of the random uncertainty is thought to be this order of magnitude.

In order to estimate the random uncertainty more accurately, we introduced the random uncertainty so as to be proportional to the half-life and lead the reduced chi-square to unity in the fitting. The total error of each data point was treated as a quadratic sum of the statistical and random uncertainties. After this procedure, the random uncertainty for each data point was estimated to be 0.23 and 0.12% for ^{196}Au and 0.17% for ^{202}Tl . The half-lives were obtained to be 6.1448 ± 0.0024 (tot.) d and 6.1451 ± 0.0012 (tot.) d from the 333 and 356-keV γ rays of ^{196}Au , respectively. Since the difference between them become much smaller, the half-life value of 6.1451 ± 0.0013 (tot.) d was determined for ^{196}Au using the both γ rays. The half-life of ^{202}Tl was determined to be 12.4706 ± 0.0055 (tot.) d. It should be noted that these errors contain both statistical and the other random uncertainties.

In the left panel of Fig. 3a, the result of the present work for ^{196}Au (closed circle) is compared to the other experimental data, open triangle [12], open square [13], open circle [14], closed-inverted triangle [15], open-inverted triangle [16], closed triangle [17], and closed square [3]. Our result of 6.1451 ± 0.0013 (tot.) d is 0.35% shorter than the recommended value of ENSDF [1].

In the left panel of Fig. 3b, the result for ^{202}Tl is compared to the other experimental data, open star [18], closed star [19], open diamond [20], closed diamond [21], and asterisk [2]. The result of this work agrees well with the latest measurement of Ref. [2].

4. Conclusion

In the present work, the half-lives of ^{196}Au and ^{202}Tl were precisely determined. The random uncertainties were estimated by the repeated measurements using HPGe detectors. The largest uncertainties are due to the random error of the order of 0.1% for each data point. The half-life of ^{196}Au was determined to be 6.1451 ± 0.0013 (tot.) d, which is 0.35% shorter than the latest value of Ref. [3]. The half-life of ^{202}Tl was determined to be 12.4706 ± 0.0055 (tot.) d with

a 93% smaller uncertainty than that of the latest value of Ref. [2].

Our measurement system can be used for the investigations on half-life changes, although it yields a random uncertainty of the order of 0.1% besides the statistical uncertainty. The random uncertainty for shorter-lived nuclides such as ^{57}Ni will be evaluated in the future plan. Further research is also needed in order to find the sources of the random uncertainty and to reduce them.

Acknowledgment. We would like to thank the accelerator and technical staff at the Research Center for Electron Photon Science for operating of the accelerator and for the other experimental supports.

References

1. Evaluated Nuclear Structure Data File (ENSDF), <http://www.nndc.bnl.gov/ensdf/>.
2. Unterwiesing, M. P.: Half-life measurements at the National Institute of Standards and Technology. *Appl. Radiat. Isot.* **56**, 125 (2002)
3. Lindenberg, K., Neumann, F., Galaviz, D., Hartmann, T., Mohr, P., Vogt, K., Volz, S., Zilges, A.: Half-lives of Au, Hg, and Pb isotopes from photoactivation. *Phys. Rev. C* **63**, 047307 (2001)
4. Ohtsuki, T., Ohno, K., Morisato, T., Mitsugashira, T., Hirose, K., Yuki, H., Kasagi, J.: Radioactive decay speedup at $T = 5$ K: electron-capture decay rate of ^7Be encapsulated in C_{60} . *Phys. Rev. Lett.* **98**, 252501 (2007).
5. Bainbridge, K. T., Goldhaber, M., Willson, E.: Influence of the chemical state on the lifetime of an isomer. *Phys. Rev.* **84**, 1260 (1951)
6. Bainbridge, K. T., Goldhaber, M., Willson, E.: Influence of the chemical state on the lifetime of a nuclear isomer, $\text{Tc}^{99\text{m}}$. *Phys. Rev.* **90**, 430 (1953)
7. Byers, D. H., Stump, R.: Low-temperature influence on the technetium-99m lifetime. *Phys. Rev.* **112**, 77 (1958)
8. Porter, R. A., McMillan, W. G.: Effect of compression on the decay rate of $^{99\text{m}}\text{Tc}$ metal. *Phys. Rev.* **117**, 795 (1960)
9. Mazaki, H., Nagatomo, T., Shimizu, S.: Effect of pressure on the decay constant of $^{99\text{m}}\text{Tc}$. *Phys. Rev. C* **5**, 1718 (1972)
10. Nishi, M., Shimizu, S.: Change in the decay constant of $^{99\text{m}}\text{Tc}$ in BaTiO_3 by the ferroelectric transition. *Phys. Rev. B* **5**, 3218 (1972)
11. Mazaki, H., Kakiuchi, S., Shimizu, S.: Search for superconducting effect on the decay of $^{99\text{m}}\text{Tc}$. *Z. Phys. B* **29**, 285 (1978)
12. Lingeman, E. W. A., Lobner, K. E. G., Nijgh, G. J., Wapstra, A. H.: The negaton transition in ^{196}Au . *Nucl. Phys.* **31**, 584 (1962)
13. Wapstra, A. H., Jansen, J. F. W., Goudsmit, P. F. A., Oberski, J.: The decay of ^{196}Au . *Nucl. Phys.* **31**, 575 (1962)
14. Bonner, N. A., Goishi, W., Hutchin, W. H., Iddings, G. M., Tewes, H. A.: Half-lives of Tm^{167} , Tm^{168} , Tm^{170} , Lu^{171} , Lu^{172} , Lu^{173} , Lu^{174} , $\text{Lu}^{174\text{m}}$, Au^{195} , Au^{196} , and $\text{Au}^{196\text{m}}$. *Phys. Rev.* **127**, 217 (1962)
15. Ikegami, H., Sugiyama, K., Yamazaki, T., Sakai, M.: Structure of platinum nuclei(I) new decay scheme of Au^{196} . *Nucl. Phys. A* **41**, 130 (1963)
16. Tilbury, R. S., Yaffe, L.: Nuclear isomers $\text{Hg}^{197\text{m}}$, $\text{Hg}^{197\text{g}}$, $\text{Hg}^{195\text{m}}$, $\text{Hg}^{195\text{g}}$, and $\text{Au}^{196\text{m}}$, $\text{Au}^{196\text{g}}$ formed by bombardment of gold with protons of energies from 8 to 60 MeV. *Can. J. Chem.* **41**, 2634 (1963)
17. Helmer, R. G.: Evaluated Decay Scheme Data for Use in the ILRR Program. ERDA/NDC-3/U, 2 (1976).
18. Hamers, H. C., Marseille, A., de Boer, Th. J.: Further experiments on the decay of ^{202}Tl . *Physica* **23**, 1056 (1957)
19. Leutz, H., Schulz, G., Wenninger, H.: Electron capture ratios in the decay of ^{202}Tl . *Nucl. Phys.* **75**, 81 (1966)
20. Cohen, I. M., Koro, R. O., Baro, G. B.: Reactor produced ^{202}Tl : determination of half-life and gamma energies. *Int. J. Appl. Radiat. Isot.* **26**, 403 (1975)
21. Taylor, H. W.: Relative γ -ray-emission probabilities in the decay of 12.6-d ^{202}Tl . *Int. J. Appl. Radiat. Isot.* **35**, 421 (1984)