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Half-life of the nuclear cosmochronometer ^{176}Lu measured with a windowless 4π solid angle scintillation detector

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The ^{176}Lu-^{176}Hf nuclear decay is a powerful tool to measure the age of astrophysical and geological events and has been used as a "cosmochronometer". However, the half-life values of ^{176}Lu measured with various experiments differ significantly. Furthermore, the half-life values evaluated from Lu-Hf isochrons in meteorites and terrestrial rocks with known ages show two different values. Here we report half-life measurements using a method that is almost independent of various uncertainties. To the best of our knowledge this is the most accurate value of ^{176}Lu half-life. We measure the total energy released from ^{176}Lu decay using a windowless 4π solid angle detector based upon bismuth germanate (BGO) scintillation crystals, where a natural Lu sample is located inside of the detector. The measured half-life of $(3.719 \pm 0.007) \times 10^{10}$ yr corresponding to a decay constant of $(1.864 \pm 0.003) \times 10^{-11}$ yr^{-1} is consistent with that obtained from the analysis of terrestrial rocks within the uncertainty.

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A meta-stable isotope ^{176}Lu decays to its daughter nucleus ^{176}Hf with a half-life of approximately $(3.5-4.1)\times10^{10}$ yr (Fig. 1). ^{176}Lu could be used as a nuclear cosmochronometer for evaluation of the duration time from a stellar nucleosynthesis event such as slow neutron capture (s-process) in asymptotic giant branch stars[1,2] or photo-disintegration reactions (γ-process) in supernovae[3] to the solar system formation. This chronometer also is a powerful tool to study formation and evolution of planetary bodies in the solar system[4–7]. Both Lu and Hf are refractory and lithophile elements, and Lu is more compatible than Hf during melting of mantle and crust, producing the new crust with low Lu/Hf ratios and the residual mantle with high Lu/Hf ratios. Therefore, the Lu-Hf chronometer is, in particular, suitable for the study of crust-mantle evolution. In fact, the Lu-Hf system has been applied to study of the crust-mantle evolution of the Earth[5,8–11], Moon[12], Mars[7,13], Vesta[14,15], which is considered to be the parent body of eucrite meteorites, and a parent asteroid of angrite meteorites[15]. Furthermore, this system has been used for the study of orogenic movements after the formation of the crust on the Earth[16].

The most important parameter for nuclear chronometers is the half-life of a parent nucleus, and the half-life of ^{176}Lu has been measured using various nuclear experimental and cosmochemical methods. ^{176}Lu decays predominantly to an excited state with a spin and parity of 6^+ on ^{176}Hf through an emission of a β-ray, and subsequently the exited state decays to the ground state of ^{176}Hf though emission of γ-rays with energies of 88, 202, 307, and 401 keV (Fig. 1). The measurement techniques could be classified into four groups: i) γ-ray measurement using a single detector[17–24], ii) γ-γ (β-γ) coincidence measurement including sum peak method[17,25–28], iii) β-ray measurement using a liquid scintillation detector[29,30], and iv) the isochron method using meteorites and terrestrial rocks with known ages[9,31–36]. However, there are two critical problems in the measured values. In 2003, a precise value of $(4.08\pm0.03)\times10^{10}$ yr was measured by the γ-γ coincidence method[27], but it is approximately 10% longer than other values except for two data[18,20] (Fig. 2). Furthermore, there is another problem in the results measured using the isochron method. The half-life values of approximately 3.72×10^{10} yr were reported by the internal isochron method for terrestrial rocks[31,32] and meteorite phosphates from the ordinary chondrite Richardton and the primitive achondrite Acapulco[33]. However, a different half-life value around 3.50×10^{10} yr was provided by whole-rock isochron method for various meteorites such as carbonaceous chondrites, ordinary chondrites, and eucrites in 2003[9], where the carbonaceous chondrites are considered to originate from asteroids located in the outer part of the asteroid belt and the others came from the inner area. This half-life value was consistent with the previously reported values for eucrites[34,35], and, later, it was reproduced by the internal isochron for a different kind of meteorite, angrite[36]. The previous nuclear experiments cannot eliminate a possibility that this shorter half-life value is more accurate (Fig. 2). To explain these two different half-life values several models have been proposed. These are inhomogeneous isotopic abundance distribution of Hf in the early solar system originating from stellar nucleosynthesis[37], secondary Lu/Hf fractionation in parent bodies of meteorites[38,39], and decay acceleration through an isomer with a half-life of 3.7 h where the isomer is excited by cosmic radiations[40,41]. At present, there are the three systematically different values (the dashed-lines in Fig. 2). Furthermore, two evaluated values of $(3.76\pm7)\times10^{10}$ yr[42] and $(3.76\pm8)\times10^{10}$ yr[43] are known, but they have large uncertainties. Even under such circumstance, the Lu-Hf chronometer has been used for the study of crust-mantle evolution of various planetary bodies such as Earth[10], Moon[12], Mars[13], and Vesta[15] and orogenic movements of the Earth[16]. However, instead of the half-life values measured by the nuclear experiments and the two evaluated values[42,43], the decay constant of 1.867×10^{-11} yr^{-1} measured by the isochron method for terrestrial rocks[32] has been used for their analyses[10,12,13,15,16].

As presented previously, there are the three systematically different values in the half-life measurements, but there is no correlation between the values and the measurement methods (Fig. 2). This indicates that when a half-life value of ^{176}Lu is precisely measured using one of the known methods it is difficult to know whether it is close to the true value. In the γ-ray measurement method i), the half-life was reported by measuring of the 202 or 307 keV γ-ray with a scintillation detector or a high-energy resolution germanium detector. The results depend on the emission probability of the γ-ray and the detection efficiency[22,23]. The emission probabilities of these γ-rays have uncertainties. The detection efficiency is usually calibrated using standard radioactive sources but their radioactivities have typically the uncertainties of a few percent. The coincidence method ii) depends on the angular correlation between two radiations and the geometrical factor such as the size of the sample. They are in general calculated using a simulation code[27,28]. The half-life measured with a liquid scintillation detector iii) depends on the uncertainty of a calibration radionuclide such as ^{3}H[30]. The isochron method iv) requires following conditions. First, the initial isotopic abundances of Hf were homogeneous among analyzed samples. Second, the elements of Hf and Lu have been closed in a sample after its formation. Third, the formation age of the sample is known using another method such as the U-Pb chronometer. Thus, it is desirable to investigate the true half-life using an accurate method being independent of the known uncertainties and the assumptions for measurements. When an accurate value close to the true half-life is measured, it also contributes to resolve the unknown mechanism leading the two different half-life values observed in meteorites.

In this paper, we report a half-life value of ^{176}Lu measured by a method that is almost insensitive to the known uncertainties and independent of the assumptions required for the isochron

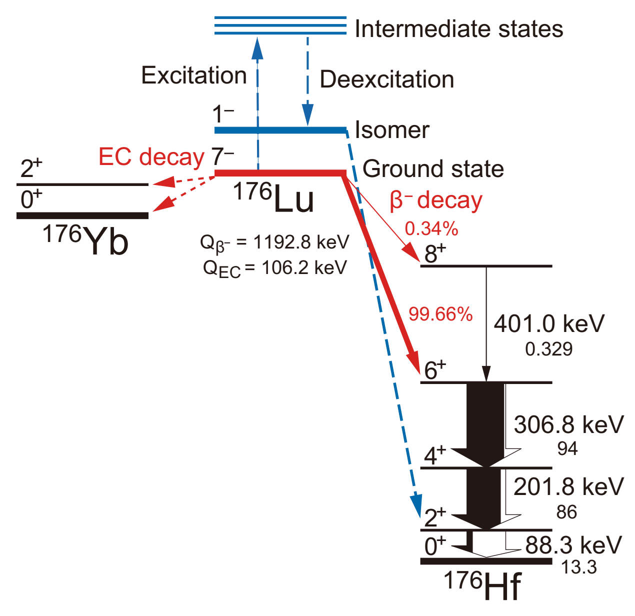

Fig. 1 Partial level scheme of ^{176}Lu and ^{176}Hf. The solid area in each arrow and the number below the transition energy indicate the emission probability of the γ ray. The blue dashed lines show decay acceleration through the isomer by an incident particle.

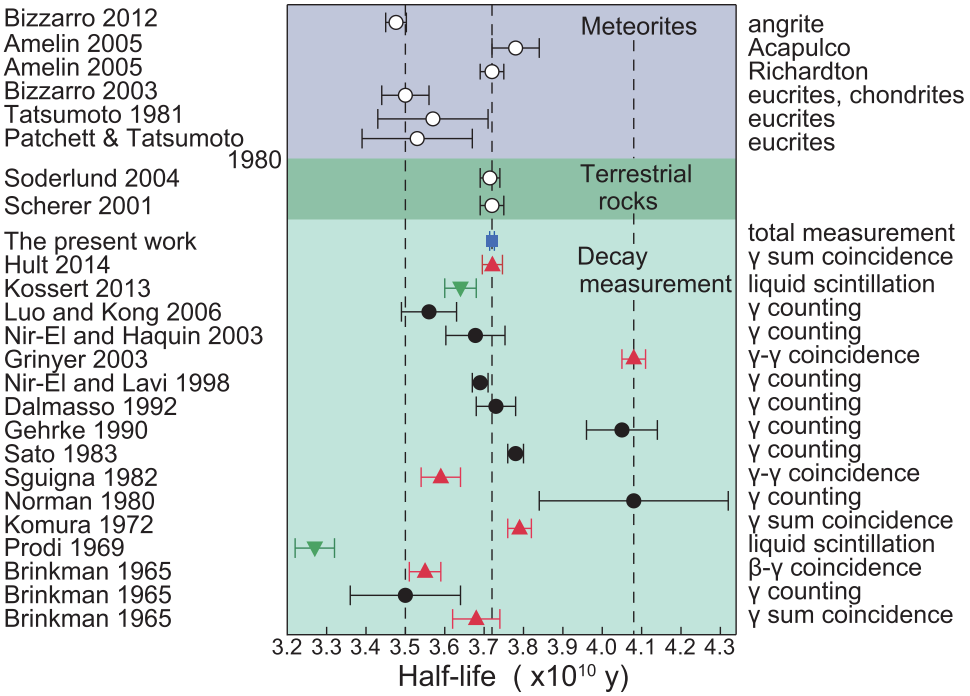

Fig. 2 Previously measured half-life values. The black filled circles indicate the half-life measurements obtained using a γ-ray detector. The red and green triangles present coincidence methods and liquid scintillation methods, respectively. The open circles show the values evaluated with the isochron methods. The three dashed lines indicate 4.08×10^{10}, 3.72×10^{10}, and 3.50×10^{10} yr, respectively. The error bars show the uncertainties taken from the original papers. Bizzarro 2012 was evaluated from its original data[36] by Iizuka et al. [39].

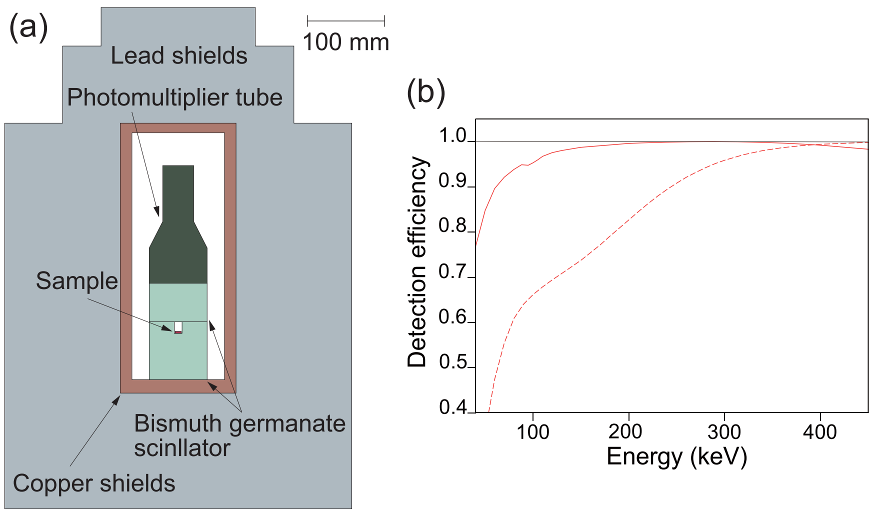

Fig. 3 Detector system. a Schematic view of experimental setup. **b** Calculated detection probabilities from radiation from the Lu sample, in which a photon or electron deposits an energy higher than or equal to 30 keV in the bismuth germanate crystals. The solid and dashed-lines indicates photons and electrons, respectively.

method. This method is different from the four techniques as explained above. We measure the total energy released from the ^{176}Lu decay using a windowless 4π solid angle detector, where a natural Lu sample is located near the center of cylindrical bismuth germanate (BGO) crystals (Fig. 3) and the detection efficiency of the decay is close to 100%. This type of detectors have been used for measuring of decay of radionuclides[44]. We obtain a half-life value of $(3.719 \pm 0.007) \times 10^{10}$ yr, corresponding to a decay constant of $(1.864 \pm 0.003) \times 10^{-11}$ yr^{-1}. This result is consistent with that obtained from the isochron method for terrestrial rocks, and we need a mechanism to explain relatively short half-life value obtained from analysis for some meteorites. We discuss the possibility that decay acceleration is caused by neutrons generated from cosmic rays.

Result and discussion

Obtained half-life of ^{176}Lu. Figure 4a, b shows the energy spectra using a ^{133}Ba standard source (see Methods). Figure 4c shows one of the Lu sample spectra and the background spectrum. The background spectrum shows Pb x-rays and a 570-keV energy γ-ray, which originate from ^{207}Bi ($T_{1/2} = 31.5$ yr) contained in the BGO crystals. Natural bismuth usually includes ^{207}Bi produced by nuclear reactions on natural lead isotopes with high-energy cosmic-ray under the ground[45]. In addition, the 662-keV γ-ray is

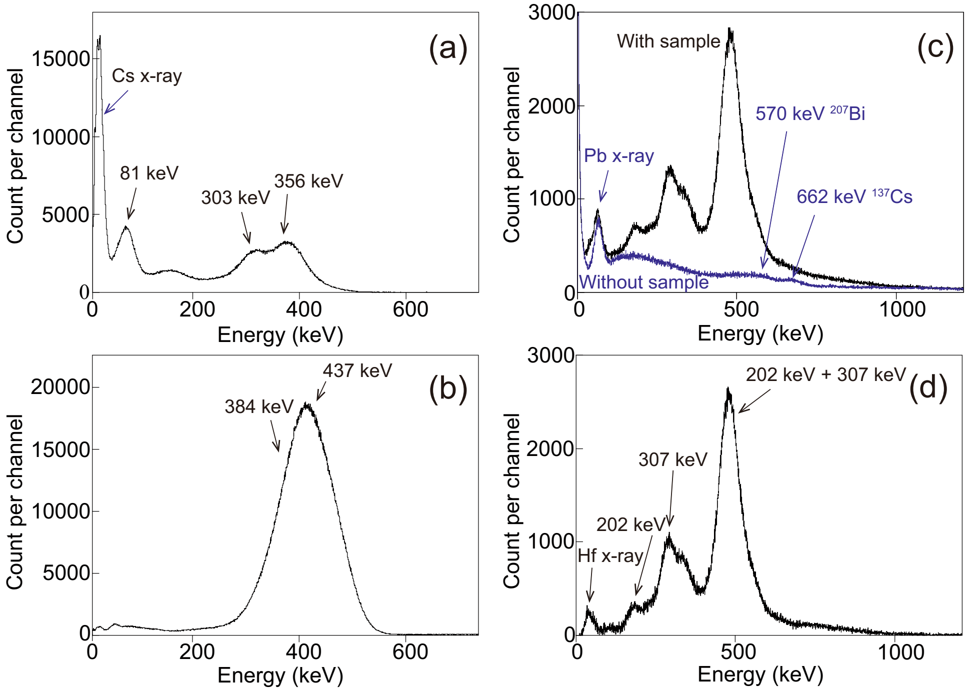

Fig. 4 Measured energy spectra using the bismuth germanate scintillation detector. a Radiation from the standard source ^{133}Ba located outside of the detector. **b** ^{133}Ba spectrum located inside of the detector. **c** Spectra from the Lu sample inside of the detector and the background without the sample. **d** Spectrum by subtracting the background from the Lu spectrum.

measured through the shields, which is radiated from ^{137}Cs contaminated inside of the experimental room. The ^{137}Cs radionuclides came from the accident of the Fukushima-Daiichi nuclear power plant in March 2011[46]. Figure 4d shows the spectrum subtracted by the background from the Lu spectrum. The 202 + 307 keV sum peak as well as Hf x-rays, 202, and 307 keV γ-rays are observed, and the continuous part in an energy range of 700–1000 keV shows the pile-up of these γ-rays and β ray. The half-life can be obtained using the following equation

$$T_{1/2} = \frac{\ln(2) \cdot t \cdot m \cdot Na \cdot \epsilon \cdot f \cdot I}{Y \cdot A}, \quad (1)$$

where t is the measuring time, m is the mass of the sample, Na is Avogadro number of 6.02214×10^{23} mol^{-1}, ϵ is the total detection efficiency, f is the elemental fraction in the sample, I is the isotopic abundance of ^{176}Lu, Y is the measured yield in an energy range of 30–1192.8 keV, and A is the atomic mass of the natural Lu of 174.967. The total counts are 1135911 ± 1467 and 1100590 ± 1455 for the samples with a mass of 125.26 ± 0.05 and 121.43 ± 0.05 mg, respectively. The measuring time is 171834 and 171832 s, respectively. The total detection efficiency is 99.9 ± 0.1% and the elemental abundance of Lu is 99.9 ± 0.1% (see Methods). The isotopic abundance of ^{176}Lu in natural samples is 2.5987 ± 0.0012%[47]. The relative uncertainties of the detection efficiency, elemental abundance, isotopic abundance, and masses are evaluated to be 0.1%, 0.1%, 0.05%, and 0.04%, respectively, and the time fluctuation is smaller than 0.01%. The uncertainties of Avogadro number and the atomic mass of Lu are negligibly small. Taking these uncertainties into account, we obtain the systematical uncertainty of 0.15%. The half-life values obtained from the two Lu samples are $[3.718 \pm 0.005(\text{stat.}) \pm 0.006(\text{sys.})] \times 10^{10}$ yr and $[3.720 \pm 0.005(\text{stat.}) \pm 0.006(\text{sys.})] \times 10^{10}$ yr. The total uncertainty is also obtained by calculating the uncertainty propagation for Eq. (1). The finally obtained half-life is $(3.719 \pm 0.007) \times 10^{10}$ yr corresponding to the decay constant of $(1.864 \pm 0.003) \times 10^{-11}$ yr^{-1}.

The present method has following four advantages. First, the obtained value is almost insensitive to nuclear structure such as γ-ray emission probability. Eq. (1) shows that the half-life depends on neither the γ-ray emission probability nor internal electron conversion coefficient. Furthermore, this detector is not sensitive to the branching ratio of the electron capture (EC) to ^{176}Yb, which has not been measured precisely. The upper limits of the EC to the ground state and the first exited state are 0.36% and 0.45%, respectively[48]. Even if the fraction of EC is not negligibly small, the present detector could measure Yb x-rays in EC. Second, the presently obtained half-life is almost insensitive to the calculated result for detection efficiency, because the detection efficiency is close to 100%. Third, the detection efficiency is free from the uncertainty of calibration sources because calibration sources have not been used. Forth, this method is free from the problem that the two different values were measured in meteorites. Thus, we conclude that the presently measured half-life is more accurate than those measured by the previous experiments as shown in Fig. 2. In addition, the present measurement is the most precise among all the methods.

The present half-life is consistent with the previous results measured by nuclear experiments[17,21–23,28] within the uncertainties. However, this is much shorter than the result measured by the γ-γ coincidence method in 2003[27] although this method has an advantage that it is not sensitive contamination of raidoactivites in samples. An array of twenty germanium detectors with Compton suppressors was used for a complex analysis of the combination of the γ-γ coincidence and the sum-peak methods. When only two γ-rays are radiated in a decay, the decay rate can be calculated by a simple formula. In contrast, when three or more γ-rays are radiated from a radionuclide, one should correct various coincidence modes such as the pile-up of three γ-rays which destroys a sum-peak event[27]. In the case of use of the twenty detectors with a combination of 380, its correction is more difficult.

The present value is also consistent with those obtained by the isochron method using terrestrial rocks with known ages determined by the U-Pb chronometer[31–33]. This indicates that the present half-life is consistent with the precisely measured half-lives of ^{235,238}U. With both chronometers the more precise ages for samples could be obtained, and even if the U-Pb chronometer cannot be used, the Lu-Hf system could be used for dating a sample consistent with the U-Pb method. Although the value reported by the isochron for terrestrial rocks[32] has been used for analysis in cosmochemistry and geochemistry at present[10,12,13,15,16], the present result shows that there is no need to change drastically the results using this value. The Lu chronometer is a powerful tool to study the ages of astrophysical events, evolution of planetary bodies in the solar system, and orogenic movements of the Earth. The present result contribute to these studies with more accurate and precise age evaluation.

Neutron induced reactions for decay acceleration of ^{176}Lu. The two different half-life values were reported using the isochron method on meteorites, but the present result shows that the longer half-life value measured in the ordinary chondrite Richardton and the primitive achondrite Acapulco[31–33] is more accurate, whereas the other value from eucrites[9,34,35], some chondrites[9], and an angrite[36] is shorter than the true value. Thus, the previously proposed models may explain the shorter half-life value. The decay acceleration via cosmic radiations is one of the candidates to explain the shorter value[40,41]. It has been known that ^{176}Lu decays to ^{176}Hf through an isomer with a half-life of 3.7 h at 123 keV in high temperature stellar environments, where an intermediate state in ^{176}Lu is first excited by photon absorption and subsequently de-excites to the isomer through photon emission (the dashed lines in Fig. 1)[2,49,50]. As a strong photon source in the early solar systems, various mechanisms such as γ-rays from radionuclides such as ^{26}Al, radiation from the protosun, γ-ray bursts near the solar system, and supernova cluster around the sun were considered[40]. As an alternative radiation source, neutrinos from a core-collapse supernova near the early solar system has been proposed[41]. Thrane et al.[41] pointed out that even if the energy of an incident photon is as high as 1 MeV, it can penetrate only a few-cm depth in typical solid states. To resolve this problem it has been suggested that the decay acceleration by cosmic rays with energies of up to 100 TeV, which can penetrate 10–20 m depth[41]. From the energy balance without detailed nuclear reactions, it was presented that a supernova near the early solar system may provide cosmic-rays enough to accelerate ^{176}Lu decay[41]. If decay acceleration occurs, the abundance of ^{176}Lu relative to ^{175}Lu should decrease. However, no evidence of the ^{176}Lu deficient has been found in meteorites[10,51].

We here point out that ^{176}Hf could be produced by two neutron induced reactions simultaneously generated by spallation reactions with high-energy cosmic-rays. One is the β decay following the ^{175}Lu(n, γ)^{176}Lu^m reaction[2,49], and the other is the decay from the isomer fed by neutron inelastic scattering on ^{176}Lu. A neutron incident on ^{176}Lu may form a compound nucleus ^{177}Lu, which decays to an exited state on ^{176}Lu through neutron emission in neutron inelastic scattering and subsequently the exited state may decay to the short-lived isomer in ^{176}Lu. The effect of neutron capture was observed in the analysis of the isotopic abundances of Hf in mesosiderites, where epi-thermal neutrons in an energy range of 0.5 eV–500 keV played more important role than thermal neutrons[37]. The neutrons produced by spallation reactions with high-energy ions have typically the energies of MeV, and the neutrons lose the energies though scattering. The ^{176}Lu decay is accelerated by the inelastic scattering with neutrons with energies from 123 keV to a few MeV, whereas ^{176}Lu as well as ^{176}Lu^m are newly produced by neutron capture on ^{175}Lu with neutrons in wide energy range from thermal energy to a few MeV. Thus, this scenario is not inconsistent with the previous results that the deficient of ^{176}Lu by decay acceleration was not found in meteorites[10,51]. The neutron inelastic scattering cross sections to ^{176}Lu^m have not been measured well and its detailed study is beyond of the scope of the present work.

Methods

Sample materials. We prepare two natural Lu metallic foils supplied from Nilaco Co. Their elemental fraction of Lu is 99.9% evaluated precisely by the supplier using x-ray resonance analysis and we take its uncertainty of 0.1%. The size of the two natural Lu metal foils is approximately 8 mm × 8 mm size and 0.125 mm thickness. The masses of the two samples are 125.26 ± 0.05 and 121.43 ± 0.05 mg that are measured before the decay measurement, and the masses measured one month later are identical with these, respectively. There is a possibility that other meta-stable isotopes with half-lives of $10^8 - 10^{11}$ yr may exist in the sample and they may contribute to the decay count. Possible radionuclides are ^{40}K, ^{87}Rb, ^{138}La, ^{147}Sm, ^{187}Re, ^{232}Th, and ^{235,238}U. Thus, we measure the fractions of the elements including these meta-stable isotopes in a small area of 40 μm × 180 μm in the same material using an inductively coupled plasma mass spectrometry (ICP-MS). The results are 0.001% for K, 0.001% for Rb, 0.005% for Re, and 0.017% for Th. The fractions of La, Sm, and U are lower than the detection limit of approximately 0.001%. Because impurities are not, in general, distributed homogeneously in a sample, the fractions obtained from the small area are not the average values of the sample. Thus, this result is not the clear evidence that these samples do not include these radionuclides, but it suggests that the fractions of them may not be enough high to affect the decay rate.

Detector system. We measure the total energy of photons and electrons radiated from ^{176}Lu using a windowless 4π solid angle detector, where a natural Lu sample is located near the center of cylindrical bismuth germanate crystals (Fig. 3a). This type of detectors have been used for measuring of decay of radionuclides[44]. Even if several radiations are simultaneously detected, we count only one event. The BGO materials have the remarkable features of non-hygroscopic and large stopping power originating from the high-Z element bismuth. Thanks to the non-hygroscopic it is possible to make a windowless detector. The BGO crystals supplied from Ohyo Koken Kogyo Co., LDT. can be divided into a cylindrical crystal with a size of ϕ3 inch. × 2 inch. and a well-type crystal having ϕ3 inch. × 3 inch. size with a hole of ϕ10 mm × 15 mm. The top and bottom surfaces of the BGO crystals are polished, whereas their sides and the insides of the hole are not polished. The sides of both the crystals and the bottom surface of the well-type crystal are covered by polytetrafluoroethylene reflection tapes. One of the natural Lu samples is located on the bottom of the hole of the well-type crystal. After the sample is set in the hole, the two crystals are connected with a photomultiplier tube (PMT) (Hamamatsu Photonics K.K., R1307-07). An ortec 113 scintillation preamplifier and an ortec 672 spectroscopy amplifier are used for amplification of the PMT signal and the output of the amplifier is recorded in an APG7400A multi-channel analyzer (K. K. TechnoAP). The detector is shielded against natural backgrounds by 15-mm thickness copper plates and lead blocks with a typical thickness of 150 mm. The counting rates with/without the Lu sample are typically 12 and 4 count per second. In the present experiment, the background fluctuation may originate from that of cosmic-rays in relatively long-time scale. To avoid this effect, the decay from each Lu sample and the background without the samples is measured for 48 h, respectively.

Detector efficiency. The thickness of the BGO crystals is chosen to detect almost all γ-rays from ^{176}Lu. Figure 3b shows the detection probabilities of photons and electrons from the natural Lu sample calculated using the PHITS particle and heavy ion transport code system[52]. We set the geometry of the detector in Fig. 3 and the Lu sample as the radionuclide source where each source point is assumed to be distributed homogeneously. Thus, the self-absorption inside of the sample is taken into account. The detection of a radiation (photon or electron) means that the deposit energy of the radiation is higher than or equal to 30 keV. This lower limit is reasonable because the Cs x-ray at approximately 31 keV from a ^{133}Ba radionuclide source can be clearly observed as presented later. The detection probabilities of photons with energies of 200–350 keV are higher than 99.5% (Fig. 3b) and thus this detector is suitable for detection of 202 and 307-keV γ-rays from ^{176}Lu. The internal-conversion coefficients and the electron energies are obtained using the BrIcc calculation code[53]. The electron energies are a few-ten keV lower than the transition energy between two states. The electron detection probability decreases with decreasing electron energy because low-energy electrons may be absorbed inside of the target. The detection probabilities for the 88, 202, and 307-keV transitions taking the internal conversion into account are approximately 75.2%, 95.0%, and 99.6%, respectively. In the decay of ^{176}Lu, a β-ray with a continuous energy is radiated, and the calculated detection probability of the β rays with the expected energy spectrum from ^{176}Lu is approximately 50.8%. We conservatively evaluate the total detection probability per ^{176}Lu decay of approximately 99.9% with an uncertainty of 0.1%.

Examination using a ^{133}Ba radionuclide source. The detector could be examined using a ^{133}Ba standard source. Figure 4a, b shows the measured spectra of the outside and inside positions, respectively. When the source is placed outside the detector individual γ-rays and Cs x-rays from ^{133}Ba are observed (Fig. 4a). In contrast, in the inside the sum peaks of 437 and 384 keV are measured (Fig. 4b) because two excited states at 437.0 keV and 383.8 keV are fed with fractions of 85.4% and 14.5%, respectively, in electron capture of ^{133}Ba. Although the BGO is slightly thin for detection of ^{133}Ba decay, the yield obtained from the sum peak spectrum is consistent with the radioactivity of the source within the uncertainty.

Data availability

Most of the data in this study are available in the paper. The other data, including raw data, can be provided upon request to the corresponding author.

Code availability

The PHITS simulation code is distibuted from Japan Atomic Energy Agency. https://phits.jaea.go.jp/index.html.

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T.H. carried out the design, perform, and analysis of the main experiment and wrote the manuscript. T.S. contributed to the analysis. T.I. motivated the experiment, measured the impurities of the sample, and contributed to the discussion.

Competing interests
The authors declare no competing interests.

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