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Precise Half-Life Determination of ^{131}Ba and ^{125}Xe

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Barium-131 and Xenon-125 are valuable radioisotopes with numerous scientific and medical applications. We produced both isotopes via proton irradiation of a cesium iodide (CsI) target at the Brookhaven Tandem Van de Graaff accelerator (BTVG). Following irradiation, we conducted systematic γ-ray spectroscopy measurements at the National Nuclear Data Center (NNDC) decay station using a calibrated High-Purity Germanium (HPGe) detector, collecting data over a 2-month period (spanning approximately 4.5 half-lives for ^{131}Ba). Through careful analysis of the characteristic γ-ray emissions from both isotopes, we determined the half-life of ^{131}Ba to be 11.55(6) days, which agrees with the previously evaluated value. For ^{125}Xe, we measured a half-life of 16.56(8) hours, slightly lower than the evaluated value.

I. INTRODUCTION

^{131}Ba is a radioisotope with significant applications in nuclear medicine[1,2] and scientific research[3,4]. With a half-life of approximately 11.5 days, the emission of a strong 124 keV gamma ray this isotope offers an ideal balance between stability for handling and gamma emissions well suited for diagnostic imaging. ^{131}Ba is a promising radionuclide for Single Photon Emission Computed Tomography (SPECT) imaging purposes[1]. It also shows potential as a diagnostic match for the alpha emitters ^{223,224}Ra[5]. ^{131}Ba additionally serves as a generator for the production of ^{131}Cs, which is utilized in brachytherapy treatments for various cancers, particularly prostate cancer[2].

The decay properties of ^{131}Ba have been measured several times[6–11], and the current evaluation provides a half-life value of 11.50(6) days[12] based on a single measurement[6] published over 30 years ago. A new measurement is timely and also recommended for measurement in Ref.[13] for neutron activation applications.

^{125}Xe is also a radioisotope of considerable value in nuclear medicine and scientific research. In medical imaging, ^{125}Xe is employed in certain lung ventilation studies[14,15], where its decay properties allow for effective visualization of pulmonary function without delivering excessive radiation doses to patients. As a radioactive noble gas, ^{125}Xe offers unique properties for tracer studies in environmental science and industrial applications, where its chemical inertness combined with detectable γ radiation makes it an ideal tracer[16]. Also serves as a generator to ^{125}I- a common isotope used in brachytherapy.

The decay properties of ^{125}Xe have also been measured several times[17–23], and the current evaluation provides a half-life value of 16.9(2) hours[12] based on two measurements[17,18] published over 60 years ago. The two averaged half-life measurements show some discrepancy, and the reported error appears to be smaller than the error of either of the individual averaged measurements. A new measurement is needed to resolve this tension.

The aim of this work is to provide new, high-precision half-life measurements. To achieve this, a sample of both ^{131}Ba and ^{125}Xe was produced via proton irradiation of CsI targets. A shielded HPGe detector was used to measure the decay of ^{131}Ba and ^{125}Xe over 2 months at Brookhaven National Laboratory. The half-lives of the isotopes of interest were obtained from analysis of the time distributions of the strongest γ-ray emissions following EC decay.

II. EXPERIMENT

Two CsI targets were irradiated at the BTVG using a 28-MeV, 500-nA proton beam. The targets for the experiment were manufactured at the Center for Accelerator Target Science (CATS) at Argonne National Laboratory (ANL). In this experiment, two thin CsI targets were employed, each with an average CsI layer thickness of approximately 2.5 mg/cm^2. One target was mounted on a carbon backing with a thickness of 40 μg/cm^2, while the other was supported by a tantalum backing with a thickness of 1.5 mg/cm^2. The proton energy was chosen to optimize the reaction channel of interest while minimizing the other reaction channels. ^{131}Ba and ^{125}Xe were produced through the ^{133}Cs(p,3n)^{131}Ba and ^{127}I(p,3n)^{125}Xe reactions, respectively.

Following irradiation of the CsI targets we conducted a series of systematic γ-ray spectroscopy measurements to determine the half-lives of the produced ^{131}Ba and ^{125}Xe isotopes. Using a HPGe detector at the NNDC decay station, calibrated with ^{152}Eu and ^{133}Ba standard sources, we collected γ-ray spectra over two months. The HPGe detector was placed in-

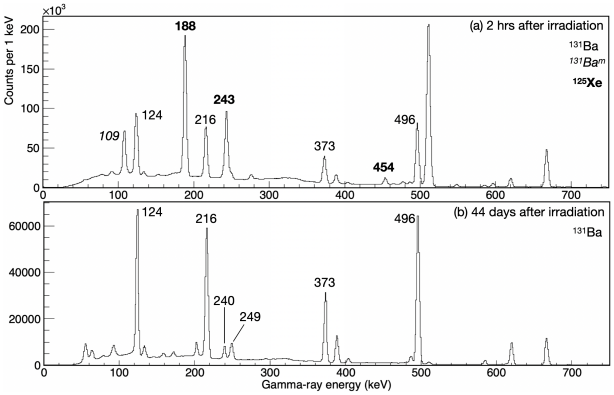

FIG. 1. A γ-ray spectrum of the CsI target after 28 MeV proton irradiation, measured using an HPGe detector (a) 2 hours and (b) 44 days following the one hour irradiation. The spectrum is shown with indicated transitions corresponding to the de-excitation of ^{125}I observed at energies of 188, 243 and 454 keV in bold while the other peaks corresponds to the de-excitation of ^{131}Cs at energies 124, 216, 240, 249, 373 and 496 keV. γ-rays from isotopes such as ^{126}I and ^{132}Cs produced via other reaction channels, were also observed.

side a graded-shield well counter which reduced the ambient background radiation by two orders of magnitude. The sample was placed inside a plastic petri dish and sealed with tape to minimize the γ-ray attenuation while ensuring integrity of the sample. The source-detector distance enabled sufficient counting statistics to be collected while keeping the dead time of the data-acquisition system to $\leq 4\%$ for each measurement. A representative γ-ray spectrum is shown in Fig. 1. The majority of the peaks in the spectrum come from the decay of ^{131}Ba and ^{125}Xe.

III. ANALYSIS AND RESULTS

Both ^{131}Ba and ^{125}Xe undergo EC decay to the excited states of their respective daughter nuclei, ^{131}Cs and ^{125}I. These excited states then de-excite by emitting characteristic γ-rays. For ^{131}Ba, we focused on its characteristic γ-ray emissions at 124, 216, and 496 keV, with relative intensities 62.0(5), 42.4(8), and 100, respectively[12] while for ^{125}Xe we monitored the prominent 188, 243 and 454 keV photo-peaks with relative intensities 100, 55.7(11), and 8.68(18)[24], respectively. Interestingly both ^{131}Ba and ^{125}Xe have $J^{\pi} = 9/2^{-}$ high spin isomers that decay to the $J^{\pi} = 1/2^{+}$ ground state through two γ-ray cascades. Half-lives of these isomers are 14.6 min and 57 s with the most prominent 109, and 111 keV γ-rays respectively[12,24]. The long-lived isomer de-excitation γ-ray is highlighted in Fig.1 (a). By gating on γ-rays followed by an EC decay, we can achieve a clean selection of the isotope of interest, allowing for a precise determination of the parent isotope's EC decay half-life.

γ-ray spectra were collected with CAEN digitizers for about a 2 month period. To obtain the peak areas for the γ ray transitions, a modified version of the Radware gf3[25] program was used which fits the spectrum with a gaussian peak shape on a linear background. The technique involves determining the counts within the peaks by integrating the spectrum above a well-defined background. The photo-peak areas were corrected for dead-time and the correction never exceeded 4%. To determine the time distribution, the corrected peak areas were divided by the live time of the measurement and plotted against the elapsed time since the experiment was started. A simple exponential-decay law was found to provide a satisfactory fit to the data.

A. ^{131}Ba

Independent values for the half-life of ^{131}Ba were obtained for the 124, 216, and 496 keV γ-ray transitions in the ^{131}Cs daughter. A reduced χ^2-minimization fit to the decay curve of the most intense 496 keV γ ray as shown in Fig.2 (c), provided a value of 11.50(8) d for ^{131}Ba where the uncertainty includes both statistical ($\pm$0.06 d) and systematic ($\pm$0.06 d) contributions combined in quadrature. The systematic uncertainty was estimated by varying the histogram bin widths ($\pm$0.010 d) as well as the beginning ($\pm$0.051 d) and end ($\pm$0.027 d) of the fit range. Gating on the 124 and 216 keV γ-ray transitions, half-lives of 11.64(10) d and 11.55(8) d respectively were found which are consistent. Consequently, we adopt a weighted average of our three measurements to arrive at a half-life of 11.55(8) d for ^{131}Ba. This measured half-life is in agreement with both the current evaluated ENSDF value of 11.50(6) d[24] and a more precise unevaluated value of 11.52(1) d reported in Ref.[10]. The most recent measurement of the ^{131}Ba half-life reported a value of 11.657(8) d[11]. However, the focus of Ref.[11] was on cross-section measurements and the details provided on the half-life measurement do not discuss whether systematic uncertainties are considered in the reported value. The half-life of ^{131}Ba measured in this work is compared to literature values in Fig. 3.

B. ^{125}Xe

The half-life of ^{125}Xe was determined by monitoring the 188 and 454 keV γ-ray transitions in the ^{125}I daughter nuclide. Although the 243 keV γ-ray transition is also a prominent emission in the ^{125}Xe decay, two contamination peaks from the ^{131}Ba to ^{131}Cs decay at energies of 240 keV and 249 keV were found to overlap with the 243 keV peak of interest. This overlap prevented accurate peak isolation and area determination for the 243 keV peak.

The half-life was determined by a reduced χ^2-minimization fit to the decay curves of the most intense γ rays as shown in Fig.4. However, the quality of the fit for the 188-keV line was poor, as evidenced by the fit residuals in Fig.4 (a). The reason for the poor quality of the fit is not understood, so only the 454-keV line was used for determining the half-life. The result is 16.56(8) h, with the systematic uncertainty estimated in the same manner as for ^{131}Ba. The systematic uncertainty was estimated by varying the histogram bin widths ($\pm$0.011 h) as well as the beginning ($\pm$0.057 h) and end ($\pm$0.020 h) of the

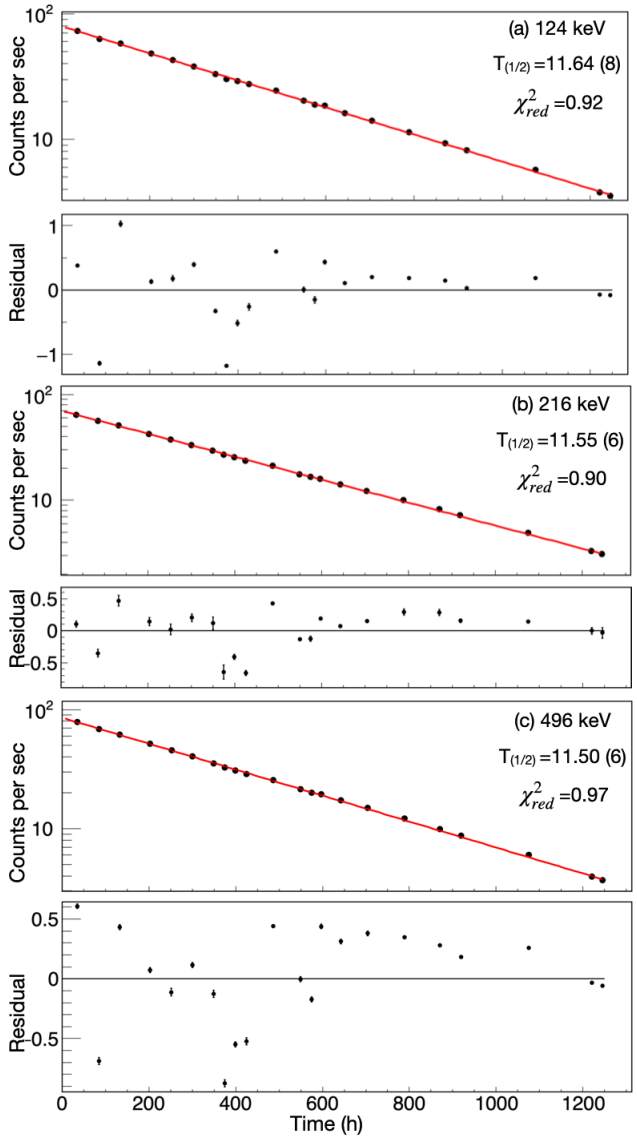

FIG. 2. Time distribution of γ-rays (a) 124 keV, (b) 216 keV, and (c) 496 keV following the decay of ^{131}Ba nuclei. The decay-time spectrum was fit with an exponential function (solid red). Only statistical uncertainties are presented in the figures. The residual is presented in the bottom panel.

fit range. This is in agreement with, although slightly shorter than, the value of 16.9(2) h currently adopted in ENSDF[24]. The present value is compared to other published results in Table I. and Fig.5.

IV. CONCLUSIONS

A new and precise measurement of the half-lives of ^{131}Ba and ^{125}Xe was performed with a sample produced by proton irradiation of CsI targets. Both half-life measurements were performed by determining the time distribution of the γ rays following the EC decay of the isotopes. ^{131}Ba half-life analysis was based on the average of the 3 most intense 124, 216, and 496 keV γ-ray transitions. The measured value of the half-life of ^{131}Ba is 11.55(8) days, which is in agreement with the half-life reported in the most recent evaluation[12]. The measured value of the half-life of ^{125}Xe is 16.56(8) hours, which is slightly lower than the half-life reported in the most recent evaluation[24].

TABLE I. Half-life obtained for ^{131}Ba and ^{125}Xe in the present work compared to literature values.

Reference	^{131}Ba Half-life (d)
1991Bo34[6]	11.50(6)
1953Co24[7]	11.8(2)
1956Be12[8]	11.52(8)
1963Ly02[9]	12.0(1)
2012Da04[10]	11.52(1)
2021KU11[11]	11.657(8)
ENSDF[12]	11.50(6)
This work, 124 keV	11.64(8)(6)
This work, 216 keV	11.55(6)(6)
This work, 496 keV	11.50(6)(6)
Reference	^{125}Xe Half-life (h)
1965An05[17]	16.8(2)
1969Lu09[18]	17.3(4)
1950An05[19]	20(1)
1952Be55[20]	18.0(4)
1960Mo09[21]	17(1)
2019SZ01[22]	16.87(8)
2021Ze01[23]	17.048(32)
ENSDF[24]	16.9(2)
This work, 188 keV	16.59(3)(6)
This work, 454 keV	16.56(6)(6)

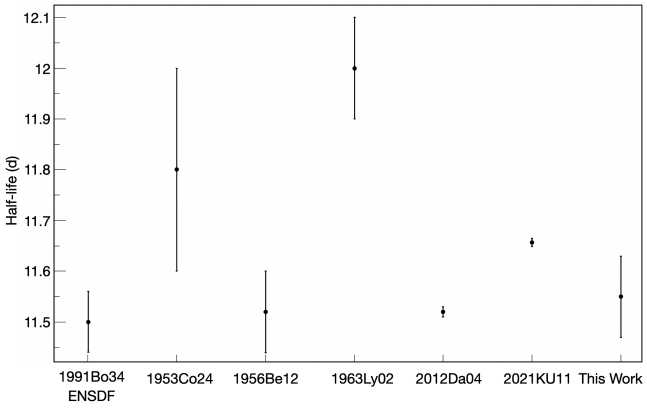

FIG. 3. Comparison of the ^{131}Ba half-life with the most recent ENSDF evaluation as well as additional results from Table I.

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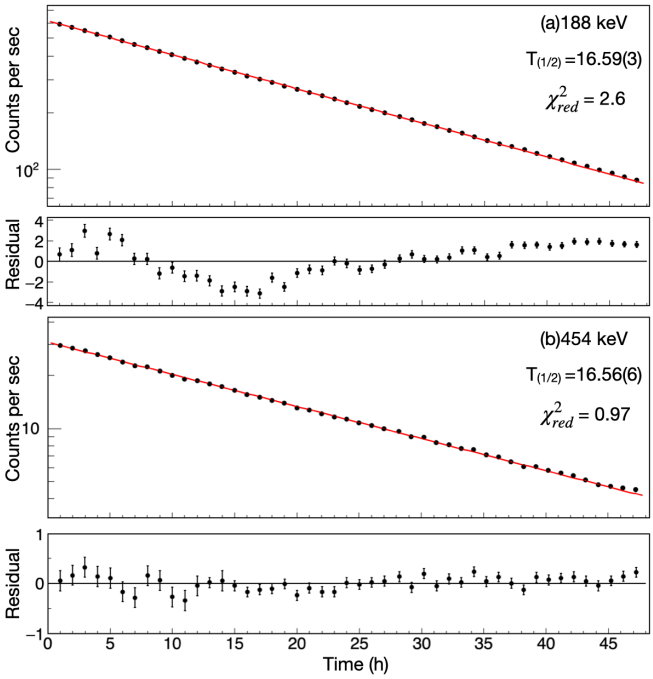

FIG. 4. Time distribution of (a)188 keV and (b)454 keV γ-rays following the decay of ^{125}Xe. The decay-time spectrum was fit with an exponential function (solid red). Only statistical uncertainties are presented in the figures. The residual is presented in the bottom panel.

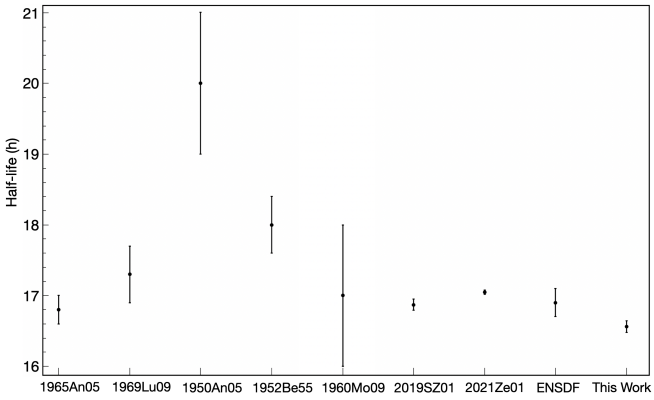

FIG. 5. Comparison of the ^{125}Xe half-life with the most recent ENSDF evaluation as well as additional results from Table I.

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