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Two determinations of the Ge-68 half-life

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HIGHLIGHTS

- The half-life of Ge-68 is determined by two methods.
- Ionization chamber measurements give a value of 271.07(12) d.
- NaI(Tl) well counter measurements give a value of 271.14(15) d.
- Both values are consistent with the DDEP recommended value and other recent determinations.
- Our value would change the DDEP recommended half-life from 270.95(26) d to 271.05(8) d.

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ABSTRACT

In nuclear medicine, ^{68}Ge is used to generate ^{68}Ga for imaging by positron emission tomography (PET) and sealed sources containing ^{68}Ge/^{68}Ga in equilibrium have been adopted as long-lived calibration surrogates for the more common PET nuclide, ^{18}F. We prepared several ^{68}Ge sources for measurement on a NaI(Tl) well counter and a pressurized ionization chamber, following their decay for 110 weeks ($\approx$ 2.8 half-lives). We determined values for the ^{68}Ge half-life of $T_{1/2}$ = 271.14(15) d and $T_{1/2}$ = 271.07(12) d from the NaI(Tl) well counter and ionization chamber measurements, respectively. These are in accord with the current Decay Data Evaluation Project (DDEP) recommended value of $T_{1/2}$ = 270.95(26) d and we discuss the expected impact of our measurements on this value.

1. Introduction

Germanium-68 decays by 100% electron capture to form ^{68}Ga, a short-lived positron emitter (Bé et al., 2013). As such, ^{68}Ge has been used for some time to make generators of ^{68}Ga for use in positron emission tomography (PET) studies. More recently, ^{68}Ge has been finding increasing use as a calibration surrogate for devices used to make industrial and clinical measurements of short-lived positron emitters (Lockhart et al., 2011; Doot et al., 2014; Zimmerman and Cessna, 2010; Zimmerman et al., 2015). A long-lived calibration source must have a well-known activity at the time of measurement. As the period of service goes on for any given source, the decay correction and its associated uncertainty grows. For any technologically important radionuclide, a good half-life with well-characterized uncertainty is crucial. In this work, we add data to a sparse set in order to increase confidence in the ^{68}Ge half-life and its uncertainty.

We report here on two determinations of the ^{68}Ge half-life by pressurized ionization chamber measurements and by NaI(Tl) well counter measurements. We carried out measurements over a period of 110 weeks ($\approx$ 2.8 half-lives). The data were acquired and analyzed in a manner consistent with other recent half-life determinations at the National Institute of Standards and Technology (NIST; Bergeron and Fitzgerald, 2015; Pibida et al., 2017), looking beyond fit uncertainties to realize conservative but realistic final uncertainties. Finally, we consider our determinations in the context of earlier work, reporting a new evaluation of the ^{68}Ge half-life, $T_{1/2}$ = 271.05(8) d.[1]

2. Materials and methods

2.1. Ge-68 sources

All ^{68}Ge sources were prepared in standard NIST borosilicate flame-sealed ampoules, with nominally 5 mL total volume. The carrier solution contained approximately 65 µg each of Ge^{4+} and Ga^{3+} in 0.5 mol L^{-1} HCl. Germanium-68 sources prepared with this carrier solution have been shown to be stable against precipitation and transfer losses for more than 7 a, or nearly 10 half-lives (Zimmerman et al., 2016). Two ampoules with initial ^{68}Ge activities of approximately

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[1] Unless explicitly stated otherwise, all uncertainties reported herein are given as combined standard uncertainties with k = 1.

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1.0 kBq (referred to as ampoules 1 and 2) and one with approximately 0.5 kBq (referred to as ampoule 3) were prepared for half-life measurements on the Wizard 2480 NaI(Tl) well counter referred to herein as "GWC" (PerkinElmer, Waltham, MA).[2] One ampoule with initial ^{68}Ge activity of approximately 3.14 MBq was devoted to half-life measurements on a pressurized ionization chamber designated "ICB" (Fitzgerald, 2010). The response from both techniques is mostly from the decay of the short-lived ^{68}Ga daughter, but all half-life measurements were performed long after parent/daughter equilibrium was established. Under these conditions, the combined parent-daughter decay curve takes the shape of a single exponential of which the slope is determined by the decay constant of the parent nuclide. Analysis by high-purity germanium spectroscopy revealed no photon-emitting impurities.

2.2. NaI(Tl) well counter measurements

The three ampoules were counted weekly on the NaI(Tl) well counter (GWC) for 75 min each, initially acquiring approximately 3 million counts in an open window (nominally 18–2000 keV) for the 1 kBq ampoules. The ampoules were centered by custom-built nylon sleeves in the typical scintillation vial sample cassettes. Between ampoules 1 and 2, an ampoule containing cold carrier solution was inserted and between ampoules 2 and 3, an empty sleeve. The average count rate for the cold carrier (4.3(1) s^{-1}) was systematically higher than for the empty sleeve (4.1(1) s^{-1}), possibly due more to rack positioning than to any actual contributions from the carrier. The cold carrier was positioned between the two 1.0 kBq sources, whereas the empty sleeve was between a 1 kBq and a 0.5 kBq source. For the half-life calculations, in each measurement cycle, the average of the cold carrier and empty sleeve count rates was used to calculate the background-subtracted count rate for all samples.

The sample activities were selected to fall in a range of GWC response linearity previously established by measuring a decaying ^{18}F source. As discussed below, response non-linearity over this range was evaluated conservatively and used to estimate an uncertainty component. Over this range, the GWC software handles deadtime corrections very well; the linearity uncertainty component includes any uncorrected deadtime effects.

No weeks were missed over the course of the experiment, but occasionally, the acquisition day varied by up to 2 d. On three occasions, elevated backgrounds in the laboratory due to parallel activities required the rejection of a count. Further, the sources were measured several times in the first three days, so that ultimately, n = 116, 115, and 114 counts were used for ampoules 1, 2, and 3, respectively.

The long-term stability of the GWC response was monitored by counting an ^{129}I ($T_{1/2}$ = 16.1(7) × 10^6 a; Bé et al., 2004) source in each measurement cycle. Over the course of this half-life determination, the relative standard deviation on the decay-corrected count rate for the ^{129}I check source was 0.06%. As described below, this variance was propagated to estimate the single-measurement uncertainty in a manner consistent with our previous reports (Bergeron and Fitzgerald, 2015). Some of the variance in the ^{129}I response can be attributed to seasonal fluctuations in the ambient conditions in the laboratory. The room housing the GWC typically has higher temperature and relative humidity in the summer than in the winter. As discussed below, seasonal variances were carefully considered in the half-life evaluation and in the uncertainty estimate.

The sensitivity of NaI(Tl) detector responses to ambient conditions is well-understood (see, e.g., Knoll, 2000; Ianakiev et al., 2009). As has been shown clearly and repeatedly, seasonal variations in ambient laboratory conditions should not be interpreted as evidence of new physics (see, e.g., Kossert and Nähle, 2014; Kossert and Nähle, 2015; Schrader, 2016; Pommé et al., 2016).

2.3. Pressurized ionization chamber measurements

One ^{68}Ge ampoule was measured approximately weekly on ICB for over two years with some occasional missed weeks. The acquisition software ended counts either when a set standard deviation of the mean (0.02%) or a set maximum time was reached. The maximum count times were set to 300 s initially and increased to 1000 s approximately halfway through the measurements. The shortest count time was 58 s. Each measurement of the ^{68}Ge ampoule was followed by measurements of a ^{226}Ra reference source (RRS50) and of the background. The maximum count times for the radium reference source were set to 300 s, allowing the 0.02% statistical limit to be consistently achieved. Background current measurements were collected for 1000 s. For the earliest and latest measurements, the ^{68}Ge and RRS50 measurements were repeated for 3–5 insertions. Due to change in personnel, for a period in the middle of the experiment (from day 364 to day 647), there were no repeat measurements. In principle, using the net ratio of the currents produced by the ^{68}Ge ampoule to RRS50 compensates for any environmental effects on the chamber response; in practice, agreement between the half-life determined from the ratio matched very well with the value determined with net currents (vide infra). Over the course of this half-life determination, the relative standard deviation on the decay-corrected ($T_{1/2}$ = 1600(7) a; Bé et al., 2008) current was 0.06%.

3. Results and discussion

3.1. NaI(Tl) well counter

Open window counting data were analyzed as described previously (Bergeron and Fitzgerald, 2015). The half-life was determined from the net count rate data using a weighted least squares fit to a single exponential function. The uncertainty used for weighting was estimated by combining components for the counting statistics ($\sigma_s = N^{-0.5}$ where N is the total net counts), the background uncertainty (σ_b, based on the standard deviation on the average of the count rates for the cold carrier and empty sleeve), and an additional uncertainty component to account for unobserved effects (σ_x, added as necessary to achieve $\chi^2/\nu \le 1$). Table 1 gives the details of these components. The "cross-talk correction" applied in some of our experiments was not necessary in this case. The fit uncertainty, u_{fit}, calculated from the fit residuals (Fig. 1a) for each source is also given in Table 1.

The fit residuals in Fig. 1a hint at a variance component with annual periodicity, especially when smoothed by a 10 point moving average (Fig. 1b). The periodicity is more pronounced for the stronger ^{129}I check source than for the ^{68}Ge sources, and indeed autocorrelation plots for the three ^{68}Ge ampoules do not reveal any clear trend. From the amplitude of the oscillation in the ^{129}I data and Eq. (1) (Pommé et al., 2008)

$$\frac{\sigma T_{1/2}}{T_{1/2}} \approx \left\{ \frac{2}{\lambda T} \sqrt{\frac{2}{n+1}} \right\} \frac{\sigma(R)}{R}, \tag{1}$$

Table 1

Uncertainty components for weighting GWC data and uncertainty on the weighted least squares fit for each source.

	σ_s/%	σ_b/%	σ_x/%	**u_{fit}/%**
ampoule 1	0.06–0.13	0.01–0.07	0.057	**0.008**
ampoule 2	0.06–0.13	0.01–0.07	0	**0.007**
ampoule 3	0.08–0.17	0.03–0.14	0.070	**0.011**

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[2] Certain commercial equipment, instruments, or materials are identified in this paper to foster understanding. Such identification does not imply recommendation by the National Institute of Standards and Technology, nor does it imply that the materials identified are necessarily the best available for the purpose.