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Measurements of the ^{82}Sr half-life

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ABSTRACT

Half-life of ^{82}Sr was measured at the National Institute of Standards and Technology (NIST) using gamma-ray spectrometry and a $4\pi\gamma$ pressurized ionization chamber. The ^{82}Sr half-life was found to be 25.36 ± 0.03 days ($k = 1$) according to gamma-ray spectrometry and 25.34 ± 0.02 days ($k = 1$) according to the $4\pi\gamma$ pressurized ionization chamber measurements.

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1. Introduction

Activity of ^{82}Sr solutions is routinely measured at the National Institute of Standards and Technology (NIST) using gamma-ray spectrometers to provide traceability to source manufacturers. The ^{82}Sr–^{82}Rb generator system is being used in cardiovascular diagnostics. Therefore, accurate activity and impurity measurements of commercial sources are required. As the published ^{82}Sr half-life values range widely (Grant et al., 1978; Litz et al., 1953; Kruger and Sugarman, 1953; Woods et. al., 1987; Hoppes et al., 1987), it is difficult to compare results of activity measurements performed by the different entities (Table 1). At present, the Evaluated Nuclear Structure Data File (ENSDF) of the National Nuclear Data Center lists the ^{82}Sr half-life as 25.55 ± 0.15 days (Tuli, 2003), and this value does not include results by Grant et al. (1978), Woods et. al. (1987), and Hoppes et al. (1987). This paper describes ^{82}Sr half-life measurements performed at NIST using gamma-ray spectrometry and a pressurized ionization chamber.

2. Experimental setup

The ^{82}Sr used in the experiment was dispensed into two 5 mL borosilicate-glass flame-sealed ampoules with a uniform wall thickness of (0.60 ± 0.04) mm and a body diameter of (16.5 ± 0.5) mm, referred to as NIST ampoules. The source used for the pressurized ionization chamber measurements consisted of a 1 mol/L HCl solution, which contained about 80 kBq ^{82}Sr in equilibrium with its daughter ^{82}Rb and about 170 kBq ^{85}S (reference time 4 October 2007 00:00 AM EST); it was labeled as source No. 1951. The source used for the gamma-ray spectrometry measurements consisted of the same solution, but contained approximately 7.8 MBq ^{82}Sr spiked with approximately 285 kBq ^{137}Cs (reference time 12 October 2007 6:00:00 AM EST); it was labeled as source No. 1952.

One closed-end coaxial high-purity germanium (HPGe) detector, labeled as "T-detector", was used for the gamma-ray spectrometry measurements. This detector was set up according to the ANSI standard N42.14 (ANSI N42.14-1999). The detector was shielded with lead bricks, 10 cm thick, lined with 3.175 mm cadmium and 0.5 mm copper sheets. The characteristics of the detector are given in Table 2. In these measurements, the distance, h, between the center of the ampoule and the face of the detector (i.e., source-to-detector distance) was 42.6 cm. The ^{82}Sr half life determination using this system was based on 25 measurements of ampoule No. 1952 ($\approx$1-day long measurement, once a week) made during the period from 12 October 2007 to 21 May 2008.

The pressurized ionization chamber used for these measurements was a new automated $4\pi\gamma$ pressurized ionization chamber setup, called "AUTOIC". The pre-existing NIST pressurized ionization chamber, called "Chamber A", has been used for over 40 years to measure half lives and to maintain calibration factors from primary standardizations of γ-ray emitting radionuclides (Unterweger et al., 1992). Both instruments rely on a Centronic[1]

[1] Certain Commercial equipment, instruments, and materials are identified in this paper to foster understanding. Such identification does not imply recommendation or endorsement by NIST, nor does it imply that the materials and/or equipment are the best available for the purpose.

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Table 1
Summary of measured half-life values for ^{82}Sr. Uncertainties are given with a coverage factor $k = 1$.

Reference	$T_{1/2}$ (days)
Present work gamma spectrometry	25.36±0.03
Present work ionization chamber	25.34±0.02
Woods et. al. (1987)	25.342±0.053
Hoppes et. al. (1987)[a]	25.36±0.80
Grant et. al. (1978)	25.55±0.15
Kruger and Sugarman (1953)	25.5±0.5
Litz et. al. (1953)	27±NA

[a] In this publication, uncertainty ±0.03 is reported in the abstract, while a value ±0.08 is quoted as a result of calculation of the uncertainty budget for $T_{1/2} = 25.36$ days. Therefore, the latter is used in this table.

Table 2
Characteristics of the HPGe detector used in this work.

Detector parameters	T-detector
Detector diameter	55±0.1 mm
Detector length	49±0.1 mm
End cap window material	Aluminum
Window thickness	1.5±0.05 mm
Crystal-window distance	5 mm±0.5 mm
Crystal material	Germanium
Crystal hole depth	34.5±0.5 mm
Crystal hole diameter	7.5±0.5 mm
Detector side cap thickness	1.5±0.1 mm
Detector side cap material	Aluminum
Detector type	p-type
Calibration geometries	NIST ampoules

Uncertainties are given with a coverage factor $k = 1$.

IG11 reentrant ionization chamber, but, for the AUTOIC, the current is measured with a Keithley 6517A electrometer, (Keithley Instruments, Inc., Cleveland, OH, USA), and the samples are changed by a custom designed automatic sample changer (Changer Labs, Knoxville, TN, USA).

The measurement method was similar to that used in earlier half-life determinations with Chamber A (Unterweger et al., 1992), with some modifications due to the use of the new electrometer. About once per day, three items were measured in the AUTOIC: ampoule No. 1951; radium reference standard RRS1000; and a blank sample holder. Since the radium reference standard has a long half life (1600 years), it was used to monitor the response of the AUTOIC over the duration of the experiment. Thus, the ratio of the background-corrected No. 1951 current to the background-corrected and decay-corrected RRS1000 currents, R_i, was used for the half-life determination. For each measurement, the selected item was inserted into AUTOIC by a robot, and the current was recorded with the electrometer. After each insertion, current readings were taken by the electrometer quasi-continuously (duty cycle around 30%) for a period of 300–500 s. The standard deviation of the mean of the current readings for ampoule No. 1951 over that period ranged from 0.007% in October 2007 to 0.03% in April 2008, due to the decreased activity. With the pressurized ionization chamber, the half-life of ^{82}Sr, was determined from 158 measurements made during the period from 4 October 2007 to 4 April 2008.

It has been reported that half-life measurements with a commercial electrometer are sensitive to discontinuities in the instrument response in the process of range changes (Schrader, 2004). To mitigate problems due to range changing, we have measured ampoule No. 1951 twice per day. The electrometer was set to auto range for the first measurement and to the 2 nA range for the second. With this arrangement, any discontinuity upon the range change (from 2 nA to 200 pA) would be apparent. In fact, after correcting the 200 pA-range data for the previously-measured range-to-range calibration factors, the two datasets are consistent. Nonetheless, the uncertainty in the linearity and zero-offset of the electrometer, determined by previous sensitivity tests, was included in the overall uncertainty of the half-life determination.

3. Results and discussion

3.1. $4\pi\gamma$ pressurized ionization chamber measurements

As stated above, Ampoule No. 1951 contained both ^{82}Sr (in equilibrium with its daughter ^{82}Rb) and ^{85}Sr. Since AUTOIC measures the total ionization current, its response is proportional to a linear combination of the activities all the three radionuclides. ^{82}Sr and ^{82}Rb were in radioactive equilibrium, and their activities decreased with the same, ^{82}Sr, half-life, $T_{^{82}\mathrm{Sr}}$. The presence of the longer-lived ^{85}Sr added complexity to the decay curve, but also allowed ^{82}Sr to be followed for 7 half-lives, as the total ionization chamber current decreased only by a factor of 14 during this period. From a fit to the half-life data, the ratio of the response of AUTOIC to the $T_{^{82}\mathrm{Sr}}$ component of the total current to that of the $T_{^{85}\mathrm{Sr}}$ component was 2.5 at the reference time.

The ratio of the (decay corrected) net source to net RRS1000 AUTOIC response, R, is given by

$$R(t) \propto A_{^{82}\mathrm{Sr},0}\left(\varepsilon_{^{82}\mathrm{Sr}}\cdot e^{-\lambda_{^{82}\mathrm{Sr}}\cdot t} + \varepsilon_{^{82}\mathrm{Rb}}\cdot\frac{\lambda_{^{82}\mathrm{Rb}}}{\lambda_{^{82}\mathrm{Rb}}-\lambda_{^{82}\mathrm{Sr}}}\cdot(e^{-\lambda_{^{82}\mathrm{Sr}}\cdot t}-e^{-\lambda_{^{82}\mathrm{Rb}}\cdot t})\right) + A_{^{85}\mathrm{Sr},0}\cdot\varepsilon_{^{85}\mathrm{Sr}}\cdot e^{-\lambda_{^{85}\mathrm{Sr}}\cdot t} \quad (1)$$

where $A_{^{82}\mathrm{Sr,o}}$ and $A_{^{85}\mathrm{Sr,o}}$ are the initial activities of ^{82}Sr and ^{85}Sr and ε values are the response functions of the ionization chamber to the various radionuclides decays. The λ values are the decay constants, e.g., $\lambda_{^{82}\mathrm{Sr}} = \ln(2)/T_{^{82}\mathrm{Sr}}$. As $e^{-\lambda_{^{82}\mathrm{Rbr}}\cdot t}/e^{-\lambda_{^{82}\mathrm{Sr}}\cdot t}$ is less than 10^{-340}, the term $e^{-\lambda_{\mathrm{Rb}}\cdot t}$ was neglected. The equation used for the half-life determination was

$$R(t) = X\cdot e^{-\lambda_{^{82}\mathrm{Sr}}\cdot t} + Y\cdot e^{-\lambda_{^{85}\mathrm{Sr}}\cdot t} \quad (2)$$

where X, $\lambda_{^{82}\mathrm{Sr}}$ and Y were the fit parameters. Since $\lambda_{^{85}\mathrm{Sr}}$ is well known, it was fixed for the fit. The weights for the fit were defined as $w_i = s_i^2$, where s_i is the estimate of the random standard deviation for a given R_i; it was determined by

$$S_i = \sqrt{S_{s,i}^2 + S_{ins}^2} \quad (3)$$

Here, $s_{s,i}$ is the standard deviation of the mean in R_i due to the standard deviations in the means of the ampoule No. 1951, RRS1000 and background readings, while s_{ins} is the random variation due to positioning of the source when it is inserted into the chamber by the robot. This quantity was determined in previous experiments and amounted to 0.017% of the current reading.

The non-linear least-squares fit of the $N = 158$ values of R_i was carried out using the Dataplot program developed at NIST (Filliben, 1981). The fit returned a value and standard deviation of $T_{^{82}\mathrm{Sr}} = 25.339 \pm 0.004$ days with a chi-squared per degree of freedom value (χ^2/ν) of 1.014. The average absolute residual value from the fit was 0.018%, as shown in Fig. 1. The validity of the uncertainty returned by the fit depends on the assumption, among others, that the deviations of the data from the model are randomly distributed. One check of this assumption was the normal probability plot of the percent residuals (*RES*). This plot appeared qualitatively linear, indicating that the *RES*'s were normally distributed. The normal probability plot correlation

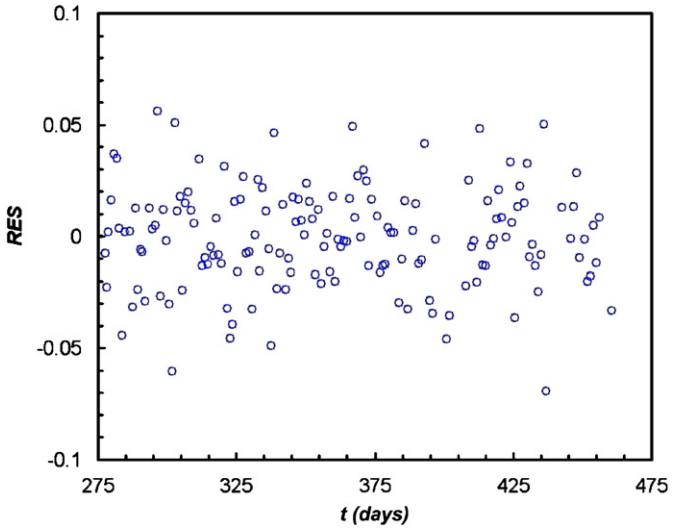

Fig. 1. Normal probability plot of the percent residuals (*RES*). The percent residuals, *RES*, were obtained by fitting $R(t)$ data using Eq. (2). All data were taken in the same range of the electrometer. The average absolute value of *RES* was 0.018%.

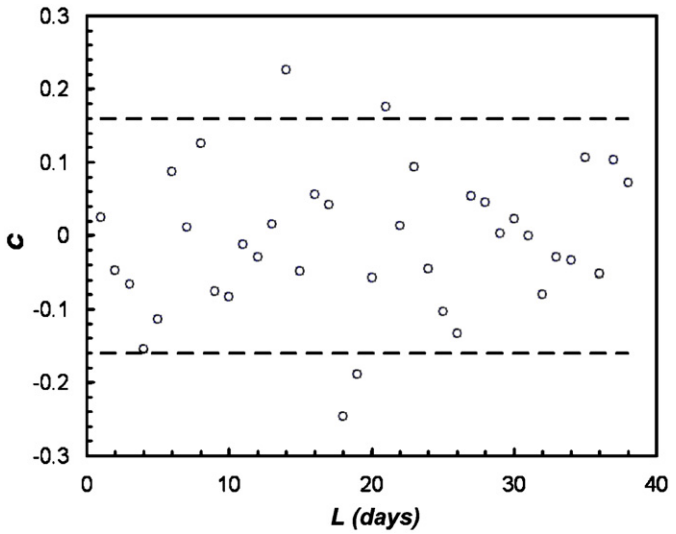

Fig. 2. Plot of the autocorrelation of the percent residuals (*RES*). Data were taken approximately once per day; so lag value, L corresponds to about one day. The dashed lines represent the 95% significance range.

coefficient derived from this plot, 0.9977, was above the tabulated critical value at the 5% significance level (0.9914) (Devaney, 1997; Filliben, 1975; NIST, 2006); so, we accept the normality hypothesis at that level. Another important test for randomness, beyond overall normality, was the autocorrelation plot shown in Fig. 2. For each lag value, L, the average correlation between each RES_i and RES_{i+L} is plotted for L ranging from 1 to $N/4$. If, for instance, near-term correlations occur, they would appear as an upturn in the plot for low L values. No such effect is evident in Fig. 2. Also, the 95% confidence interval is represented by dashed lines in Fig. 2. Since 11% of the points are outside this region, as opposed to 5% expected, the assumption of randomness is not supported by that test. Yet, there does not appear to be a general trend in the plot, nor correlations above 0.5; so, no strong conclusions can be made. It may be noted that the two high values on the plot occur at lags of 14 and 21, which may indicate a weekly trend. However, this evidence is unconvincing.

During the experiment, RRS1000 was removed from its sample holder a few times to be used in other measurement systems. This may have led to a dataset with more than one characteristic random distribution, which could introduce an additional component of variance in the value of $T_{^{82}Sr}$. Furthermore, the linearity of the response of this system is less certain than the linearity of the response of Chamber A, and the long-term stability of the small zero-offset (dark current) of the higher electrometer ranges is not yet well known. To determine the uncertainty in the half-life due to these sources, sensitivity tests were performed by modifying the data and observing the resultant half-life value from the fit. To determine the sensitivity to electrometer range changes, two sets of data were fitted. The discrepancy between the half lives resulted from these fits was 0.03%, and neither the χ^2/ν value, nor the returned uncertainty were significantly different. To test how any non-linearity in the response of the system would affect the calculated half life, the data was artificially mapped to $R_i' = R_i(1-0.001 \cdot R_i)$. At this level of induced non-linearity, which was consistent with the experimental upper limits determined by comparing the AUTOIC with Chamber A previously, the χ^2/ν value and the uncertainty in the half-life parameter were again unchanged, but the half life returned by the fit was changed by 0.05%, or 3.3 times the fit uncertainty. This test illustrates the fact that the use of a model has to be justified on the basis of the prior knowledge that it is correct, rather than purely on fit statistics. To put it differently, there is more than one model that will fit the data equally well, but only one is based on the correct physics and will, therefore, deduce the desired quantity. A similar test was carried out by introducing a small offset to the data and gauging the response. Here, the resulting half life value changed by 0.06%, while the uncertainty and χ^2/ν were nearly unchanged. In this case, some increase in the autocorrelation for low L was evident; it set a limit on the possible size of this effect. The long-term stability and characteristics of AUTOIC is still under investigation. As a result, the uncertainties due to the limits of our knowledge of the linearity and range offsets of the electrometer were included in the uncertainty of the half life determination. As shown in Table 3, the combined uncertainty, u_c, was 0.079%. Therefore, the expanded uncertainty, $U = k \cdot u_c$, was 0.16% for a coverage factor $k = 2$.

For the pressurized ionization chamber measurements, the final value for the half life of ^{82}Sr using the AUTOIC was $T_{^{82}Sr} = 25.34 \pm 0.02$ days (the interval is for $k = 1$).

3.2. HPGe measurements

In the HPGe detector measurements (Source No. 1952), the ratio of the net gamma-ray emission rates of the 776.517 keV ^{82}Sr gamma-ray line to the 661.657 keV ^{137}Cs gamma-ray line, R', was followed for 222 days (Fig. 3). The source was spiked with ^{137}Cs to monitor instrument fluctuations during the measurement time. Cesium-137 was chosen because it has a well-known long (relative to ^{82}Sr) half-life of 10976 ± 30 days (Laboratorie National Henry Bequerel, Recommended Data, 2008; Browne and Tuli, 2007) (1 year = 365.2422 days). A (25.36 ± 0.03)-day half-life of ^{82}Sr was obtained from the first-order exponential decay fit of the ratio with two different fitting programs, a χ^2-minimization routine and Origin 6.0.

With Origin, a non-weighted fit of the ratio of the net gamma-ray emission rates, R', was performed using Eq. (4):

$$R' = A_1 e^{-t/t_1}, \tag{4}$$

where A_1 is the amplitude and t_1 the decay constant (σ_{A1} and σ_{t1} are their uncertainties). Therefore, $T_{^{82}Sr}$ can be calculated from

Table 3
Evaluation of the uncertainty of the half-life of ^{82}Sr ($T_{^{85}Sr}$) determined with the automated ionization chamber (AUTOIC).

Component	Comments	Assessment type[a]	u_i (%)
Data fit	Standard deviation of fit parameter for ^{82}Sr half-life with $N = 158$ data points covering 7.5 ^{82}Sr half-lives using 3 fit parameters and $\chi^2/\nu = 1.01$.	A	0.016
^{85}Sr half life	Standard uncertainty in ^{82}Sr half life determination due to uncertainty in published ^{85}Sr half-life (64.850 ± 0.007) d (Schönfeld, E. and Dersch, R., 2004) value.	B	0.011
Model assumptions	Standard uncertainty in ^{82}Sr half life due to uncertainty in assumptions of linearity and absolute zero for current readings from apparatus, determined by sensitivity tests.	B	0.077
Relative combined standard uncertainty (u_c)			0.079
Relative expanded uncertainty (U) ($k = 2$)			0.16

[a] Assessment type "A" denotes evaluation by statistical methods and B" denotes evaluation by other methods.

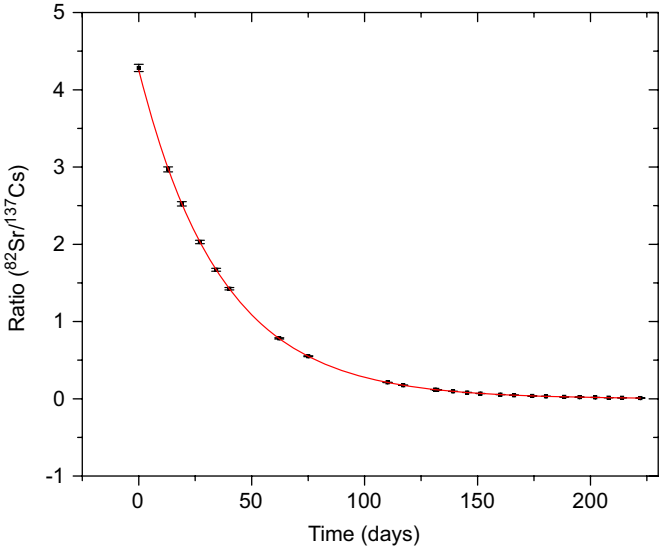

Fig. 3. HPGe measurements of the activity of the ^{82}Sr source No. 1952 in time. R is the ratio of the net gamma-ray emission rate of the 776.517 keV ^{82}Sr gamma-ray to the corresponding value of the 661.657 keV ^{137}Cs gamma-ray line.

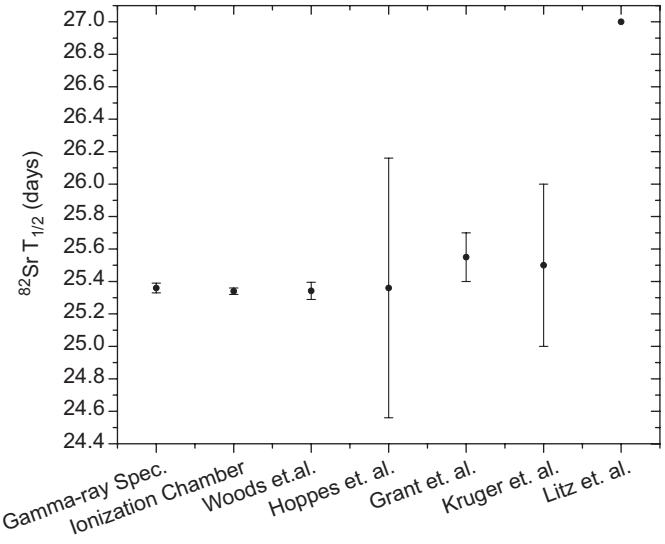

Fig. 4. ^{82}Sr half-life values determined in this and the previous works.

Eq. (5) as

$$T_{^{82}Sr} = \frac{\ln(2) t_1 T_{^{137}Cs}}{T_{^{137}Cs} + \ln(2) t_1}. \quad (5)$$

The uncertainty can be obtained by propagation of uncertainties in Eq. (6):

$$\sigma_{T_{^{82}Sr}} = \ln(2) \sqrt{\frac{(T_{^{137}Cs}(T_{^{137}Cs} + t_1 \ln(2)) - t_1 \ln(2) T_{^{137}Cs})^2}{(T_{1/2^{137}Cs} + \ln(2) t_1)^2} \sigma_{t_1}^2 + \frac{(t_1(T_{^{137}Cs} + t_1 \ln(2)) - t_1 T_{^{137}Cs})^2}{(T_{^{137}Cs} + \ln(2) t_1)^2} \sigma_{T_{^{137}Cs}}^1}. \quad (6)$$

Both fits for R' are good. The value of t_1 obtained from the Origin-produced non-weighted fit was equal to 36.677 ± 0.045 days, with a chi-squared (χ^2) value equal to 0.203 and a correlation coefficient of 0.99996. The χ^2-minimization routine yielded the chi-squared value equal to 4.6974 with a chi-squared per degree of freedom value (χ^2/ν) of 0.4519 ($\nu = 23$).

Table 1 and Fig. 4 summarize the results of our measurements and those available in the literature. The values for the ^{82}Sr half-life quoted by Woods et al. (1987) and Judge et al. (1987) are identical; so, only one of the two references is given. The values obtained in this work agree within their stated uncertainties with each other and with the results by Woods et al. and Hoppes et al. These values do not agree with the value presently quoted by the ENSDF National Nuclear Data Center of 25.55 ± 0.15 days, which corresponds to the half-life reported by Grant et al. (1978) and Tuli (2003).

4. Conclusions

The ^{82}Sr half-life was determined to be 25.36 ± 0.03 days ($k = 1$) according to gamma-ray spectrometry and 25.34 ± 0.02 days ($k = 1$) according to pressurized ionization chamber measurements. Caution should be exercised in using a half-life value in activity measurements until these new results are taken into account in the next revision of the ENSDF values.

References

ANSI N42.14-1999 American National Standard for Calibration and Use of Germanium Spectrometers for the Measurement of Gamma-ray Emission Rates of Radionuclides.
Browne, E., Tuli, J.K., 2007. Nuclear Data Sheets for $A = 137$. vol. 108, 2173–2318.

Devaney, J., 1997. Equation discovery through global self-referenced geometric intervals and machine learning. Ph.D Thesis, George Mason University, Fairfax, VA.
ENSDF, National Nuclear Data Center, Brookhaven National Laboratory 〈http://www.nndc.bnl.gov/〉.
Filliben, J.J., 1975. The probability plot correlation coefficient test for normality. Technometrics 17, 111–117.
Filliben, J.J., 1981. DATAPLOT—an interactive high-level language for graphics, non-linear fitting, data analysis, and mathematics. Comput. Gr. 15, 199–213.
Grant, P.M., Erdal, B.R., Whipple, R.E., Daniels, R.J., O'Brian Jr., H.A., 1978. Half-lives of ^{82}Sr and ^{82}Rb. Phys. Rev. C 10 (6), 2799–2800.
Hoppes, D.D., Coursey, B.M., Schima, F.J., Yang, D., 1987. National Bureau of Standards decay-scheme investigations of strontium-82–rubidium-82. Appl. Radiat. Isot. 38 (3), 195–203.
Judge, S.M., Privitera, A.M., Woods, M.J., 1987. The half-life of ^{82}Sr. Appl. Radiat. Isot. 38 (3), 193–194.
Kruger, P., Sugarman, N., 1953. Evidence for a 25.5-day Sr-76-Sec Rb chain. Phys. Rev. 90, 158–159.
Laboratorie National Henry Bequerel, Recommended Data, 19 May 2008 〈http://www.nucleide.org/DDEP_WG/DDEPdata.htm〉.
Litz, L.M., Ring, S.A., Balkwell, W.R., 1953. The 1.25-minute Rb^{82m} daughter of 27-day Sr^{82}. Phys. Rev. 92 (2), 288–290.
NIST, 2006. NIST/SEMATECH e-Handbook of Statistical Methods 〈http://www.itl.nist.gov/div898/handbook/〉 (accessed 14 May 2008).
Schrader, H., 2004. Half-life measurements with ionization chambers—a study of systematic effects and results. Appl. Radiat. Isot. 60, 317–323.
Tuli, J.K., 2003. Nuclear Data Sheets for $A = 82$. vol. 98, pp. 292–309.
Unterweger, M.P., Hoppes, D.D., Schima, F.J., 1992. New and revised half-life measurements results. Nucl. Instrum. Meth. Phys. Res. A 312, 349–352.
Woods, M.J., Judge, S.M., Lucas, S.E.M., 1987. The half-life of ^{82}Rb. Appl. Radiat. Isot. 38 (3), 191–192.