The half-life of ^{76}As

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In the course of making high-accuracy measurements of arsenic, we found that the most recently published and compiled half-life of ^{76}As did not agree with our data as well as the earlier accepted value. To redetermine this parameter, ^{76}As sources were measured on four Ge gamma detector systems, and an exponential function was fitted to the decay data by two different nonlinear least-squares methods. We obtained $T_{1/2} = 1.09379$ days with a standard uncertainty of 0.00045 days. This result is 1.5% higher than the most recent value, but is in agreement with the older, less precise, consensus value.

Introduction

In the pursuit of high-accuracy measurements of arsenic in silicon by instrumental neutron activation analysis,[1] we found that the most recently published[2] and compiled[3] half-life of ^{76}As ($T_{1/2} = 1.0778\pm0.0020$ d) did not agree with our data as well as the earlier recommended value of 1.097±0.003 d.[4–6] We therefore sought to remeasure this half-life more accurately.

Experimental

After several preliminary experiments, freshly prepared pure sources of ^{76}As were counted repeatedly and continually for a preset live time of 5000 or 10000 seconds at 20 cm from three different Ge gamma-ray detectors, along with a precision pulser.[7] The decay was followed for as long as 8 half-lives, acquiring up to $28\cdot10^6$ net counts in as many as 150 spectra. Two more sets of measurements were performed by counting other sources on two detectors for a preset clock time. In all, four detectors (ranging in relative efficiency from 26% to 45%) were used, with four amplifiers of two different models, three models of analog to digital converter (one Wilkinson and two successive-approximations), and four multichannel analyzers (MCAs) of two different types. Integral counting rates ranged from 800 to 5000 counts/s, corresponding to dead times from 12% to 1.5%. Pulse pileup rejection circuitry was not generally used, because its sensitivity to noise can sometimes lead to overcorrection in practice, in addition to some distortion of statistics.[8] One experiment did use this feature, but the half-life obtained in this case was indistinguishable from the other measurements.

For the best-characterized "Kris" detection system, the centroid of the 556 keV peak was stable: the standard deviation was 0.09 channels (0.006%) in 150 spectra collected over a range of 11.6% to 0.16% dead time in 8.7 days. In separate experiments, the pulser frequency was shown to be constant to 0.03% (s.d. of mean, $n = 11$ in 24 hours) when compared with a ^{137}Cs source. Few of these tests revealed detectable bias, and only the pulser stability contributed significant uncertainty to the final results. The livetime clock of the MCA overcorrected slightly, giving an apparent counting rate of the 25-Hz pulser 0.11% greater than that measured directly with a precision frequency counter. Numerical experiments verified that this inaccuracy did not influence the value of the half-life. The realtime clock was 0.0035% slow. Because the MCA clock was synchronized with the computer clock at the beginning of each spectrum acquisition, the more accurate computer clock (0.0009% fast relative to Universal Time) marked the decay time at which each count began.

Two computational methods were used to derive the half-life from the counting data. In the first method, three data sets (with livetime fixed) were analyzed by a straightforward nonlinear least-squares method. The net areas of the 559+564-keV doublet of ^{76}As and the pulser peak were integrated in each spectrum, using fixed channels for the peak and baseline regions.[9] A spreadsheet was used to correct the data for decay during the extended live time and for pulse pileup, and then to solve the exponential decay equation for the mean life $\tau = t_{1/2}/\ln(2)$. The procedure minimizes the weighted sum of squared residuals

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$$Q(\tau)=\sum_{i=1}^{n}\frac{\left(N_i-A(\tau)e^{-t_i/\tau}\right)^2}{V_i} \qquad (1)$$

where N_i is the corrected net peak area for measurement i beginning at decay time t_i, and $A(\tau)$ is the best-fit intercept at $t=0$. The variances V_i were computed as the sum of squares of the Poisson standard uncertainty of the observed net peak area and of the pulser correction (the latter calculated from the missing counts). The resulting uncertainty in the fitted value of τ was obtained from the parabola described by $Q(\tau)$ vs. τ: The standard uncertainty is the value of τ for which $Q=Q_{min}+1$.[10]

In the second method, all five data sets were analyzed by a novel non-linear least squares method, to be presented in detail in a separate paper, that fits a model to the gross counts in the regions of the photopeak, the pulser peak, and two regions defining the continua underlying the two peaks. Only gross counts are used in order to preserve the known Poisson distributions of the numbers of counts. In the fitting process, a Q was minimized that is similar to the Q in the fit described above:

$$Q=\sum_{i=1}^{n}\frac{(N_{s,i}-f_s(t_i))^2}{V_{s,i}}+\frac{(N_{p,i}-f_p(t_i))^2}{V_{p,i}} \qquad (2)$$

but this time, $N_{s,i}$ and $N_{p,i}$ represent gross counts in the photopeak and pulser peak regions, respectively, and the variances $V_{s,i}$ and $V_{p,i}$ were taken as equal to $f_s(t_i)$ and $f_p(t_i)$, according to the Poisson distribution and as recommended by CABELL and WILKINS,[11] with a small extra addition to take into account the variances of the gross counts in the continuum regions adjacent to the peaks. The functions f_s and f_p calculate the expected gross areas from model parameters such as the half-life of the nuclide, the peak-to-total ratio of the detector, the dead-time per pulse, the pulser frequency, and the continuum information.

Results

The results of these analyses are given in Table 1. Chi-squared per degree of freedom ranged from 0.9 to 1.4 for the first seven results. The normalized residuals $z=(N_i-A(\tau)e^{-t_i/\tau})/\sqrt{V_i}$ for experiment 4 (the longest data set) as analyzed by method 1 are plotted against decay time in Fig. 1. No trend with time is noticeable, and the distribution is approximately normal. The two fitting methods yield nearly identical results when each is applied to the first three data sets: the mean ratio is 0.9998, with the s.d. of the mean 0.02%. This agreement makes a strong case for the absence of bias through fitting, especially since the methods are so different. It cannot be said, however, that there is no bias among counting systems. Although three detectors gave concordant results in experiments 4 through 7, the peak positions in the spectra from experiment 8, the system with the oldest detector but the newest electronics, drifted downward by 0.1% as the dead time decreased from 6% to 0.1%. Although the algorithm allowed for this behavior, the half-life obtained from this detector is not concordant with the rest, and chi-squared per degree of freedom is 1.7. We therefore calculated our best value as the mean of the weighted means of experiments 4 through 7: 1.09379 days. The standard deviation of the mean is $s/\sqrt{4}=0.00031$ d, or 0.03%.

Table 1. Half-life of ^{76}As derived from five experiments

Experiment	Detector system	Method	Half-life, d	Uncertainty, d
4 LT	Kris	1	1.09383	0.00022
		2	1.09435	0.00028
5 LT	Tracy	1	1.09332	0.00035
		2	1.09332	0.00036
6 LT	Ty	1	1.09325	0.00032
		2	1.09318	0.00033
7 CT	Kris	2	1.09455	0.00027
8 CT	*Sarah*	*2*	*1.09052*	*0.00019*

LT denotes preset live time and CT preset clock time. Uncertainties are given as the standard uncertainty of the fit. Experiment 8 is discussed separately in the text.

Table 2. Uncertainty budget for ^{76}As half-life

	Source	$u(i)$	unit	DF
Type A	Measurement replication	0.00031	d	3
	Pulser stability	0.00033	d	10
Type B	Computer clock	0.00001	d	∞
	Combined standard uncertainty u_c	0.00045	d	
	Expanded uncertainty $U_c=2.0\cdot u_c$	0.00091	d	

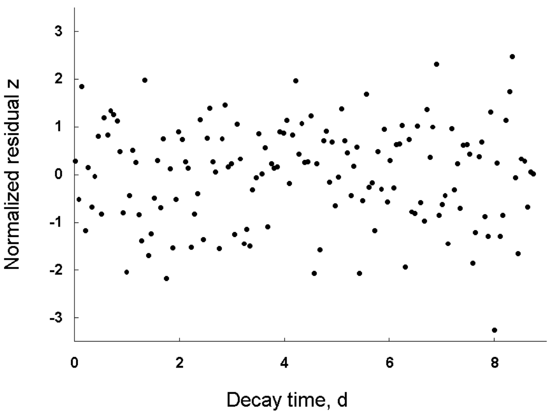

Fig. 1. Normalized residuals from a fit by method 1 to the data from experiment 4, plotted as a function of decay time. Of the 150 data points, 99 (66%) are within 1 standard uncertainty of zero and 142 (95%) within 2

The uncertainties in this measurement are collected in Table 2. With greater than 10^7 counts in each experiment, the Poisson counting uncertainty is negligible. As discussed above, the counting system used in experiments 4 and 7 had a standard error in the pulser frequency (in agreement with previous measurements)[7] of 0.033%, comparable with the dispersion among experiments. The same pulser was used in all four of the accepted measurements. The only systematic (type B) uncertainty is the computer clock.

Discussion

The value of 1.09379 d, with a combined standard uncertainty u_s = 0.00045 d, for the ^{76}As half-life obtained in these measurements is in agreement with that inferred from our INAA measurements.[1] It agrees within uncertainties with the older value of EMERY et al.,[4] which was obtained in seven measurements with six different kinds of detectors. Our value differs greatly from MIGNONSIN's recent result.[2] We tested our algorithms by measuring the well-characterized half-life of ^{198}Au, collecting 250·10^6 net counts in 83 spectra over 20 days. We obtained by both of our methods a best value of 2.6927 d with a standard uncertainty of 0.0002 d. This is within 0.1% of the value of 2.6952±0.0002 d[12] adopted in ENSDF, although the uncertainties do not overlap. However, our measurement of the ^{198}Au half-life is 0.3% greater than MIGNONSIN's value of the same quantity. In the latter paper, four half-lives are reported (of ^{198}Au, ^{24}Na, ^{76}As and ^{42}K) that are all low (by 0.4%, 0.6%, 1.6% and 3.5%, respectively) compared with earlier recommended values. A possible explanation is that this author fitted his data on a log-linear scale, a method that has been shown to give systematically short half-lives[13]and to give differing results depending on the duration of the experiment.[11] Our own results imply that the claimed uncertainty of a half-life based on measurements obtained with a single detector system should be suspect, at least until the small differences we observe are better understood.

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