



## ISTITUTO NAZIONALE DI RICERCA METROLOGICA Repository Istituzionale

A new low-uncertainty measurement of the  $^{31}\text{Si}$  half-life

This is the author's accepted version of the contribution published as:

*Original*

A new low-uncertainty measurement of the  $^{31}\text{Si}$  half-life / D'Agostino, Giancarlo; Di Luzio, M; Mana, Giovanni; Oddone, M.. - In: METROLOGIA. - ISSN 0026-1394. - 54:3(2017), pp. 410-416. [10.1088/1681-7575/aa6edf]

*Availability:*

This version is available at: 11696/55327 since: 2020-05-19T10:36:37Z

*Publisher:*

IOP

*Published*

DOI:10.1088/1681-7575/aa6edf

*Terms of use:*

This article is made available under terms and conditions as specified in the corresponding bibliographic description in the repository

*Publisher copyright*

Institute of Physics Publishing Ltd (IOP)

IOP Publishing Ltd is not responsible for any errors or omissions in this version of the manuscript or any version derived from it. The Version of Record is available online at DOI indicated above

(Article begins on next page)

# A New Low-Uncertainty Measurement of the $^{31}\text{Si}$ Half-Life

G. D'Agostino<sup>(1)</sup>, M. Di Luzio<sup>(1),(2)</sup>, G. Mana<sup>(3)</sup> and M. Oddone<sup>(2)</sup>

(1) Istituto Nazionale di Ricerca Metrologica (INRIM), Unit of Radiochemistry and Spectroscopy, c/o Department of Chemistry, University of Pavia, via Taramelli 12, 27100 Pavia, Italy

(2) Department of Chemistry, University of Pavia, via Taramelli 12, 27100 Pavia, Italy

(3) Istituto Nazionale di Ricerca Metrologica (INRIM), Strada delle Cacce 91, 10135 Torino, Italy

Email of the corresponding author: [g.dagostino@inrim.it](mailto:g.dagostino@inrim.it)

Submitted to: *Metrologia*

**Abstract:** Half-life values are widely used in nuclear chemistry to model the exponential decay of the activity of radionuclides. The analysis of existing half-life data values reveals a general lack of information on the performed experiments and, in a few cases, an almost complete absence of uncertainty budgets. This is the situation for  $^{31}\text{Si}$ , the radionuclide produced via neutron capture reaction recently used to quantify trace amounts of  $^{30}\text{Si}$  in a sample of the  $^{28}\text{Si}$ -enriched material produced for the determination of the Avogadro constant. In particular, the now recommended 157.36(26) min value is the weighted average of ten data acquired between the 40's and the 90's and published without a detailed discussion of the uncertainty budget. In order to improve the  $^{31}\text{Si}$  half-life value we carried out repeated observations of the  $^{31}\text{Si}$  decay rate via  $\gamma$ -ray spectrometry measurements. This paper reports the result we obtained, including details of the experiment and the evaluation of the uncertainty.

## 1. Introduction

A special issue of the journal *Metrologia* was recently dedicated to the evaluation of uncertainties in radionuclide metrology [1]. The measurement of the half-life of radionuclides was one of the covered topics [2]. In particular, it was reported that for many radionuclides the dispersion of the half-life values obtained in different determinations is larger than anticipated from their uncertainties. Moreover, details on the experiment and how the uncertainty was evaluated are occasionally missing.

The reliability of recommended half-life values concerns different fields, including analytical techniques based on radioactivity [3]. Since half-life correction factors are used to rescale the measured activities to a reference time, the value of the half-life may bias the result of the quantified analyte and error propagation may affect the combined uncertainty.

This was the case of a recent measurement of the  $^{30}\text{Si}$  mole fraction,  $x(^{30}\text{Si})$ , of a silicon single crystal highly enriched in  $^{28}\text{Si}$  used for the determination of the Avogadro constant [4]. The relative contribution of the adopted  $^{31}\text{Si}$   $t_{1/2}$  value [5] to the combined uncertainty of  $x(^{30}\text{Si})$  was 35%. Although the data used to derive the recommended value are not discrepant, a close check of the referenced papers confirms the lack of details on experiments and uncertainty evaluation. For example, the uncertainty of one of the most recent published data [6] was intentionally increased from 0.012 min to 0.5 min by evaluators [5] to account for systematic effects.

Hence, aiming at improving the knowledge of the  $^{31}\text{Si}$  half-life, we carried out an experiment based on neutron activation and  $\gamma$ -counting. Details of the measurement are reported in the following, including the equation model, the corrections and the uncertainty budget of the result.

## 2. Measurement model

The way we propose to measure the  $^{31}\text{Si}$  half-life,  $t_{1/2}$ , is to carry out an experiment consisting of production of  $^{31}\text{Si}$  via neutron capture reaction and observation of the exponential decay of the  $^{31}\text{Si}$  activity via  $\gamma$ -ray spectrometry. The observation is a series, hereafter called sequence, of successive count rate measurements of the 1266.1 keV  $\gamma$ -photons emitted during the radioactive decay of a  $^{31}\text{Si}$  source [2]. The experiment is repeated several times to reduce the statistical uncertainty.

The 1266.1 keV  $\gamma$ -photons count rate at the beginning of the  $i^{\text{th}}$  count of the  $j^{\text{th}}$  sequence,  $C_{ij}(t_{d\ ij})$ , starting at  $t_{d\ ij}$  and lasting  $t_{c\ ij}$ , is

$$C_{ij}(t_{d\ ij}) = \frac{\lambda n_{ij}}{(1 - e^{-\lambda t_{c\ ij}})} \delta_{ij} f_{ij}, \quad (1)$$

where  $\lambda = \ln(2)/t_{1/2}$  is the  $^{31}\text{Si}$  decay constant,  $n_{ij}$ ,  $\delta_{ij}$  and  $f_{ij}$  are the net count of the 1266.1 keV  $\gamma$ -photon peak, the dead time correction and the pile-up correction of the  $i^{\text{th}}$  count of the  $j^{\text{th}}$  sequence, respectively.

The  $n_{ij}$  value is obtained by subtracting the background count,  $b_{ij}$ , from the gross count,  $g_{ij}$ . The  $\delta_{ij}$  and  $f_{ij}$  values depend on the rate of pulses at the input gate of the  $\gamma$ -spectrometer and correct for the lost counts due to dead time and pile-up, respectively. In the case of  $\lambda t_{c\ ij} \ll 1$  and  $t_{\text{dead}\ ij} \ll t_{c\ ij}$ ,

$$\delta_{ij} = \frac{t_{c\ ij}}{t_{c\ ij} - t_{\text{dead}\ ij}} \quad \text{and} \quad f_{ij} = e^{\mu(t_{\text{dead}\ ij}/t_{c\ ij})}, \quad (2)$$

where  $t_{\text{dead}\ ij}$  is the dead time during  $t_{c\ ij}$ , i.e. the overall time the detection system spent with the input gate inhibited, and  $\mu$  is a (positive) constant, hereafter called pile-up factor, due to residual errors after compensation for count loss following leading edge pile-up. Hereafter,  $t_{\text{dead}\ ij}/t_{c\ ij}$  is defined as the relative dead time obtained by measuring in live-time correction mode and indicates the fraction of lost counts during  $t_{c\ ij}$ . In the case of a low rate of pulses,  $\rho$ , at the input gate of the  $\gamma$ -spectrometer,  $t_{\text{dead}\ ij}/t_{c\ ij}$  corresponds to  $\rho \tau$ , where  $\tau$  is the dead time per pulse [2].

In equation (2),  $t_{\text{dead}\ ij}/t_{c\ ij}$  might be replaced by  $\rho \tau$  to compute the pile-up correction as a function of  $\rho$ . However, the knowledge of  $\rho$  would require an additional rate meter connected to the electronics whereas the  $t_{\text{dead}\ ij}/t_{c\ ij}$  value is directly measured with negligible uncertainty and embedded in the  $\gamma$ -spectrum information.

The  $C_{ij}(t_{d\ ij})$  value is proportional to the  $^{31}\text{Si}$  activity, with the constant of proportionality being the detection efficiency of the  $\gamma$ -photons,  $\eta$ , which takes into account (i) the source-detector geometry, (ii) the intrinsic efficiency for the particular gamma-ray energy, (iii) the incomplete absorptions due to Compton scattering and (iv) the probability of emission of the observed gamma radiation.

If the  $j^{\text{th}}$  sequence of counts is performed without moving the source and under stability conditions of the detection system,  $\eta_j$  is constant and the variation of the count rate, equation (1), with time can be modeled by

$$C_{ij}(t_{dij}) = \hat{C}_j(t_{d1j}) e^{-\lambda t_{dij}} + \varepsilon_{ij}, \quad (3)$$

where  $\hat{C}_j(t_{d1j})$  is the expected value of the count rate at the starting time of the first count of the  $j^{\text{th}}$  sequence,  $t_{d1j}$ , and  $\varepsilon_{ij}$  is the error term, i.e. the deviation of the count rate mainly due to counting statistics. Here and hereafter, the mark (^) is used to define the expected value only in the equations used as fitting models and omitted elsewhere.

### 3. Experiment

The detection system used in this study consists of a coaxial Ge detector ORTEC<sup>®</sup> GEM50P4-83 (relative efficiency 50%, resolution 1.90 keV FWHM at 1332 keV) connected to a digital signal processor ORTEC DSPEC 502.

The rise time and flat top digital filter shaping-time constants were set to 12  $\mu\text{s}$  and 1  $\mu\text{s}$ , respectively, corresponding to a dead time per pulse,  $\tau$ , of approximately 38  $\mu\text{s}$  [7]. The acquisition was performed in extended live-time correction mode according to Gedcke-Hale method with pulse pile-up rejection in automatic set threshold; the system corrected for coincident pulses with the exception of events closely spaced in time below its fast time resolution.

Since the end cap of the detector is placed inside a low-background graded lead shield located in an underground laboratory, the input pulse rate due to background is limited to approximately 15 pulses per second. The temperature of the room is controlled at 23  $^{\circ}\text{C}$ .

The stability of the detection system was checked by recording a sequence of 750 successive counts of the 661.8 keV  $\gamma$ -photons emitted during the radioactive decay of a  $^{137}\text{Cs}$  source. The uninterrupted observation time was 375 h.

The pile-up correction factor was measured by performing several counts of the 661.8 keV  $\gamma$ -photons of the  $^{137}\text{Cs}$  source with the addition of a  $^{152}\text{Eu}$  source located at different positions with respect to the detector end cap to change the rate of pulses at the input gate of the  $\gamma$ -spectrometer [8]. Two sequences of 34 counts were repeated with the  $^{152}\text{Eu}$  source in six different positions and two sequences of 34 counts were repeated without the  $^{152}\text{Eu}$  source. The overall observation time was 320 h and consisted of 476 counts.

In both cases, the  $^{137}\text{Cs}$  source was fixed at about 60 mm from the detector end cap to keep constant the detection efficiency of the 661.8 keV  $\gamma$ -photons. In addition, the counting window,  $t_c$ , was adjusted on line to reach a 0.1% relative uncertainty of the 661.8 keV  $\gamma$ -peak net count due to counting statistics, i.e. a net count value close to  $10^6$ .

The measurement of the  $^{31}\text{Si}$  half-life was carried out with a 6 g sample of a high purity natural silicon single crystal. This sample was used as a  $^{31}\text{Si}$  source produced by the activation of  $^{30}\text{Si}$  via the neutron capture reaction  $^{30}\text{Si}(n,\gamma)^{31}\text{Si}$ . Twelve repeated neutron activations and observations of the decay rate were performed. On average, a single sequence lasted 400 min and consisted of 26 successive counts. The overall observation time was 169 h and consisted of 310 counts.

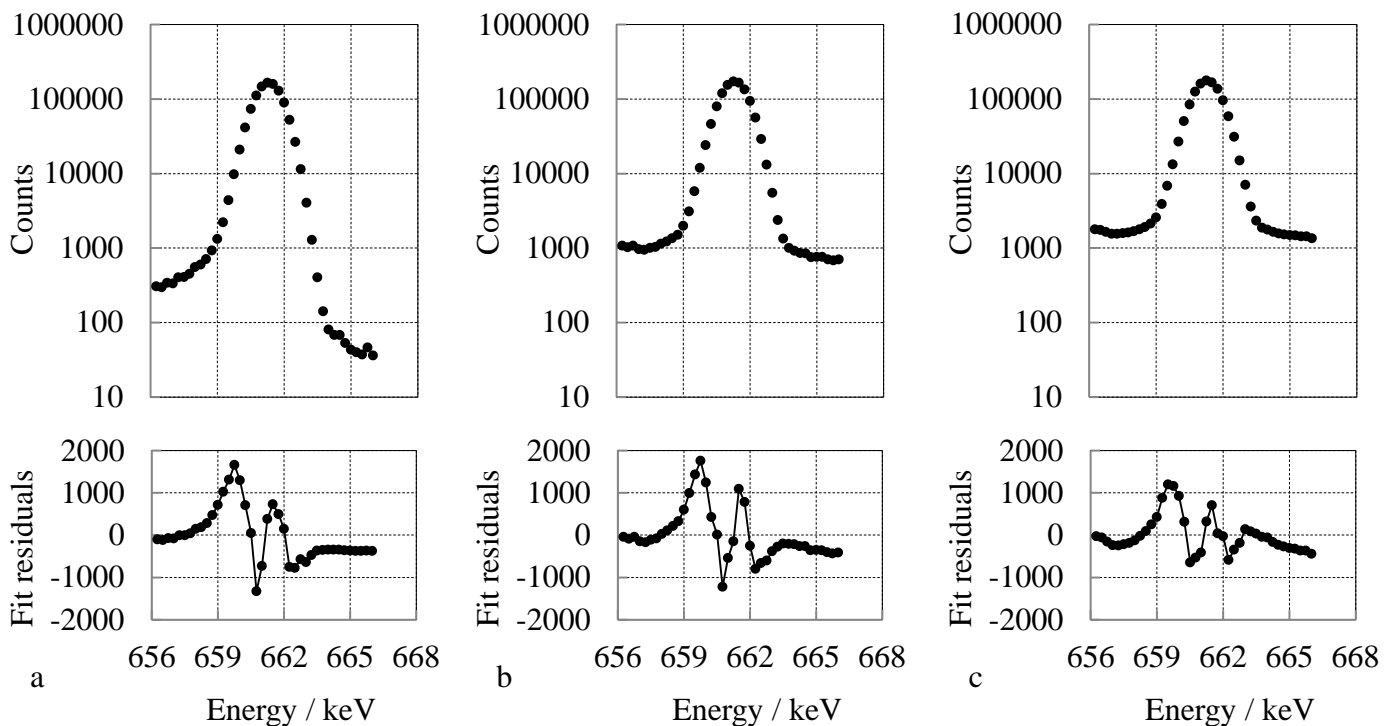
Each neutron irradiation lasted 3 h and was carried out at the central thimble of a 250 kW TRIGA Mark II reactor. After activation, the  $^{31}\text{Si}$  source was fixed at about 10 mm from the end cap of the detector. The  $^{31}\text{Si}$  decay rate was observed by recording a sequence of counts of the 1266.1 keV  $\gamma$ -

photons collected with a constant detection efficiency. The counting window was adjusted on line to achieve a 1% relative uncertainty of the 1266.1 keV  $\gamma$ -peak net count due to counting statistics, i.e. a net count value close to  $10^4$ .

#### 4. Data analysis and discussion

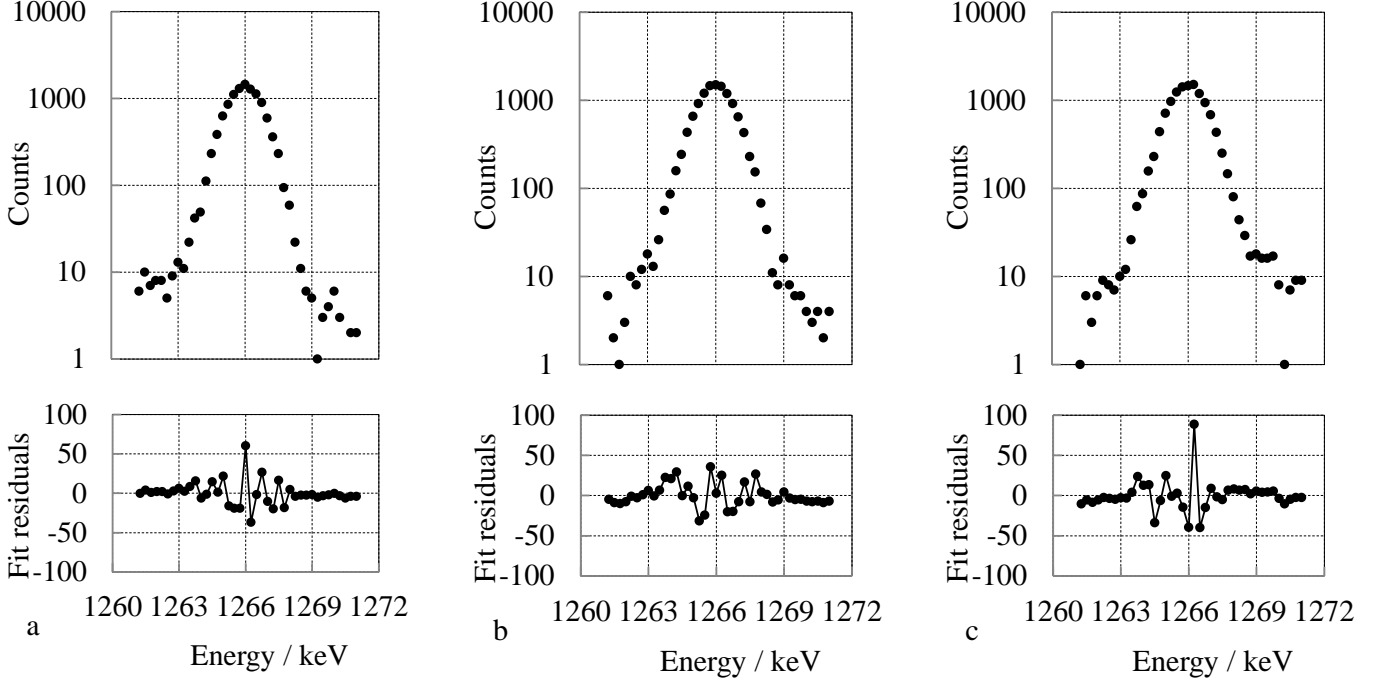
The net areas of the  $\gamma$ -peaks were estimated from the collected spectra using the WAN32 analysis algorithm of the ORTEC<sup>®</sup> Gamma Vision software [9].

The zoom of three  $\gamma$ -spectra showing the 661.8 keV  $^{137}\text{Cs}$   $\gamma$ -peak and fit residuals collected during the measurement of the pile-up correction factor is plotted in figure 1. The  $\gamma$ -peak in figure 1a, 1b and 1c refers to a relative dead time of the detection system,  $t_{\text{dead } ij}/t_{c ij}$ , of 0.077, 0.188 and 0.286, respectively.



**Figure 1.** Zoom of three  $\gamma$ -spectra collected during the measurement of the pile-up correction factor showing the 661.8 keV  $^{137}\text{Cs}$   $\gamma$ -peak and the fit residuals obtained with (a) 0.077, (b) 0.188 and (c) 0.286 relative dead time.

The zoom of three  $\gamma$ -spectra showing the 1266.1 keV  $^{31}\text{Si}$   $\gamma$ -peak and fit residuals collected during a sequence performed for the measurement of the  $^{31}\text{Si}$  half-life is plotted in figure 2. The  $\gamma$ -peak in figure 2a, 2b and 2c refers to a  $t_{\text{dead } ij}/t_{c ij}$ , of 0.079, 0.191 and 0.287, i.e. at the end, middle and start count of the sequence, respectively.



**Figure 2.** Zoom of three  $\gamma$ -spectra collected during the measurement of the  $^{31}\text{Si}$  half-life showing the 1266.1 keV  $^{31}\text{Si}$   $\gamma$ -peak and the fit residuals obtained with (a) 0.079, (b) 0.191 and (c) 0.287 relative dead time.

#### 4.1. Stability and pile-up of the detection system

The stability and pile-up factor of the detection system was checked and measured, respectively, by processing the  $\gamma$ -spectra recorded during the sequences of counts of the  $^{137}\text{Cs}$  source.

The count rate of the  $^{137}\text{Cs}$  661.8 keV  $\gamma$ -photons measured at the  $i^{\text{th}}$  count of the  $j^{\text{th}}$  sequence,  $C_{ij}(t_{dij})$ , and corrected for decay at the starting time of the first count of the first sequence,  $t_{d11}$ , is

$$C_{ij}(t_{d11}) = \frac{C_{ij}(t_{dij})}{e^{-\lambda(t_{dij}-t_{d11})}}, \quad (4)$$

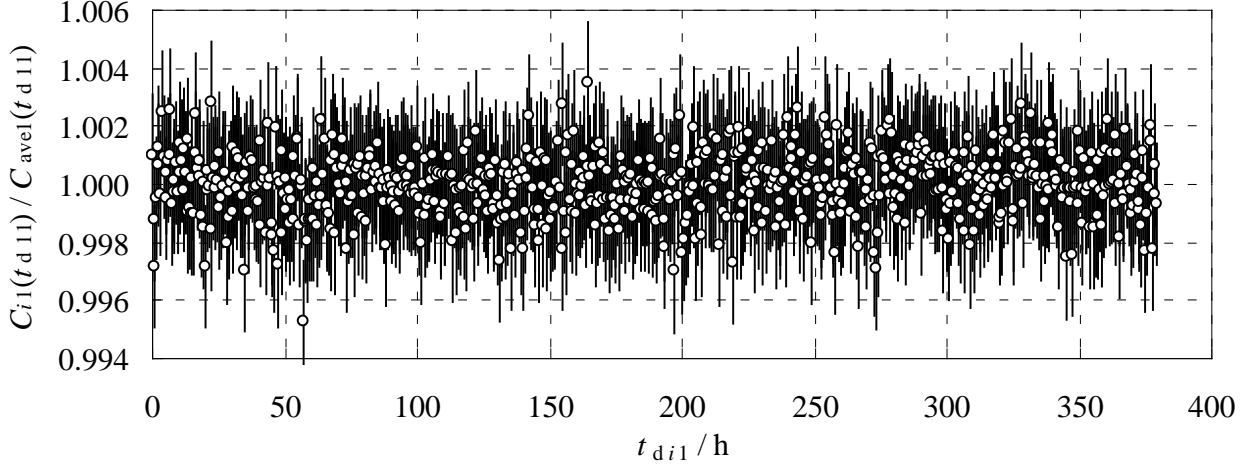
where  $C_{ij}(t_{dij})$  is obtained from (1) using the  $^{137}\text{Cs}$  decay constant,  $\lambda$  ( $t_{1/2} = 30.08$  y), and the net count of the 661.8 keV  $\gamma$ -peak,  $n_{ij}$ . Since the  $\gamma$ -photons counting was performed with a constant detection efficiency,  $C_{ij}(t_{d11})$  is constant as well.

In addition, the following model applies:

$$C_{\text{pu } ij}(t_{d11}) = \hat{C}_{ij}(t_{d11}) e^{-\mu(t_{\text{dead } ij} / t_{cij})} + \varepsilon_{ij}, \quad (5)$$

where  $\hat{C}_{ij}(t_{d11})$  is the expected value of  $C_{ij}(t_{d11})$  and  $C_{\text{pu } ij}(t_{d11}) = \frac{\lambda n_{ij}}{(1 - e^{-\lambda t_{cij}}) e^{-\lambda(t_{dij}-t_{d11})}} \delta_{ij}$  is the  $\gamma$ -photons count rate measured at the  $i^{\text{th}}$  count of the  $j^{\text{th}}$  sequence uncorrected for pile-up and corrected for decay at  $t_{d11}$ .

The single sequence of 750 counts collected for the stability test was performed at a fixed relative dead time / input pulse rate, 0.077 (i.e. 7.7% of lost counts) / 2000 s<sup>-1</sup>. Therefore,  $j = 1$  (single sequence) and  $i = 1, 2, \dots, 750$  (counts); the  $\delta_{i1}$  and  $f_{i1}$  values are constant. In the case of a stable detection system, the ratio of the  $C_{i1}(t_{d11})$  value, obtained from equation (4), to the average of the  $C_{i1}(t_{d11})$  values,  $C_{ave1}(t_{d11})$ , must vary due to counting statistics around the expected unity value without a deterministic drift. This was the case of the ratios obtained in this study and plotted in figure 3.



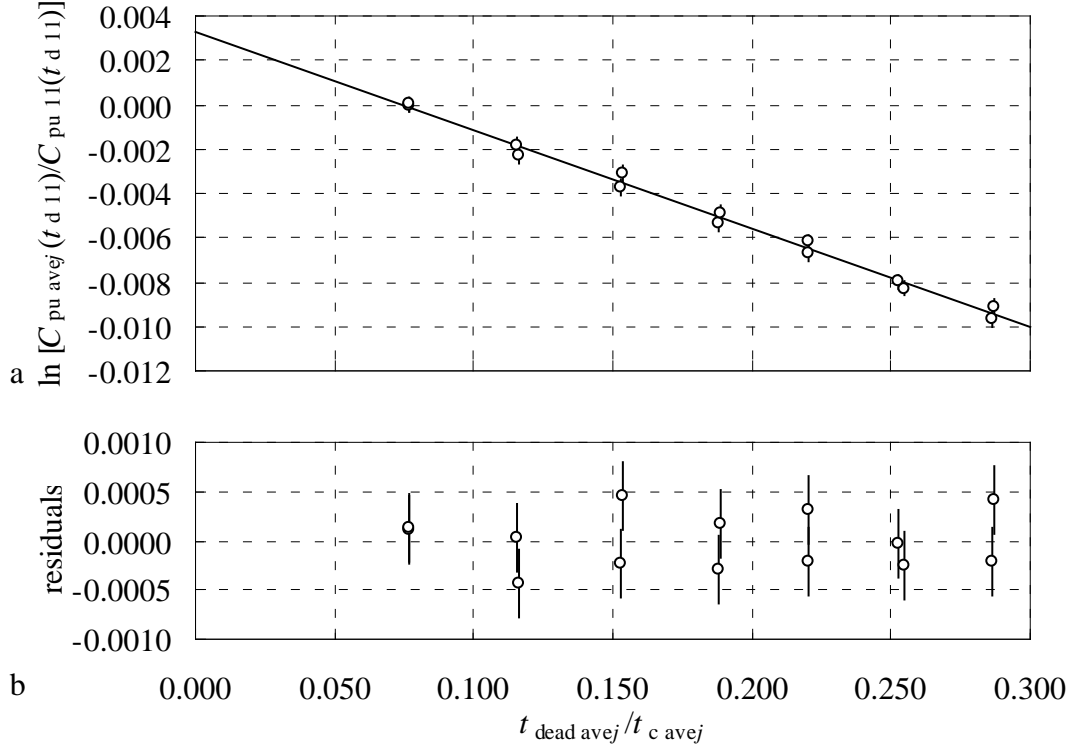
**Figure 3.** The normalized measured count rate of the <sup>137</sup>Cs source corrected for decay as a function of time. The error bars indicate a 95% confidence interval due to counting statistics.

The average relative uncertainty due to counting statistics of the 661.8 keV  $\gamma$ -peak net count and evaluated by the WAN32 analysis algorithm is  $1.048 \times 10^{-3}$  whereas the experimental standard deviation of the ratios is  $1.064 \times 10^{-3}$ . The experimental standard deviation is 1.5% higher than the expected standard deviation according to Poisson statistics. This (small) difference might be due to medium frequency instability of the detection system.

The fourteen sequences of 34 counts collected for the measurement of the pile-up factor were performed with the detection system working at seven different relative dead times / input pulse rates, i.e. 0.077 / 2000 s<sup>-1</sup> (1<sup>st</sup> and 8<sup>th</sup> sequence), 0.116 / 3000 s<sup>-1</sup> (2<sup>nd</sup> and 9<sup>th</sup> sequence), 0.153 / 4000 s<sup>-1</sup> (3<sup>rd</sup> and 10<sup>th</sup> sequence), 0.189 / 5000 s<sup>-1</sup> (4<sup>th</sup> and 11<sup>th</sup> sequence), 0.220 / 6000 s<sup>-1</sup> (5<sup>th</sup> and 12<sup>th</sup> sequence), 0.253 / 7000 s<sup>-1</sup> (6<sup>th</sup> and 13<sup>th</sup> sequence), and 0.288 / 8000 s<sup>-1</sup> (7<sup>th</sup> and 14<sup>th</sup> sequence). Therefore,  $j = 1, 2, \dots, 14$  (sequences) and  $i = 1, 2, \dots, 34$  (counts); the  $\delta_{ij}$  and  $f_{ij}$  values are changing according to equation (2).

Linear least squares regression is applied to estimate the pile-up factor,  $\mu$ , by fitting a straight-line to the natural logarithm of equation (5) averaged with respect to the 34 counts of the  $j^{\text{th}}$  sequence,  $C_{pu\ avej}(t_{d11})$ , and normalized to the  $C_{pu11}(t_{d11})$  value. Expressly, the measured variable,  $y_{ij}$ , is  $\ln[C_{pu\ avej}(t_{d11})/C_{pu11}(t_{d11})]$ , the independent variable  $x_{ij}$  is  $t_{dead\ avej}/t_{c\ avej}$  and the estimated parameters are the intercept  $\ln[\hat{C}_{ij}(t_{d11})/C_{pu11}(t_{d11})]$  and the slope  $-\mu$ . Since the main contribution to the uncertainty of  $C_{pu\ ij}(t_{d11})$  is the fixed 0.1% relative uncertainty of the 661.8 keV  $\gamma$ -peak net count, the standard deviation of the error term of  $y_{ij}$ ,  $\varepsilon_{ij}/C_{pu\ avej}(t_{d11})$ , is constant and close to  $0.001/\sqrt{34}$ .

The straight line fitting the data and the residuals are shown in figure 4a and figure 4b, respectively. The error bars confirm that the logarithm of uncertainties due to counting statistics are constant. Thus, the straight-line fitting is recommended instead of the of the exponential function fitting required in the case of non-adapted uncertainties.



**Figure 4.** The straight line (a) and the residuals (b) obtained by fitting the data collected to determine the pile-up factor. The data collected in two repeated sequences for each  $t_{\text{dead avej}}/t_{\text{c avej}}$  value are shown. The error bars indicate a 95% confidence interval due to counting statistics.

Since the residuals of three out of fourteen data are somewhat larger than the expanded uncertainty due to counting statistics, there must be an additional influence factor affecting the estimated  $4.45(8) \times 10^{-3}$  pile-up factor,  $\mu$ . Here and hereafter the uncertainties ( $k = 1$ ) in parentheses apply to the last respective digits. Furthermore, the  $\mu$  value obtained using the  $^{152}\text{Eu}$  as interfering source might be biased due to a possible energy dependence of the correction [10]. In fact, the  $^{152}\text{Eu}$   $\gamma$ -spectrum includes 23  $\gamma$ -peaks with energies between 120 keV and 1500 keV; the resulting effect is different with respect to the pile-up occurring in the case of single 1266.1 keV  $^{31}\text{Si}$   $\gamma$ -peak counting.

It may be observed that the residuals confirm the application of the pile-up correction defined in (2) at least up to relative dead times / input count rates close to  $0.300 / 8300 \text{ s}^{-1}$ . In addition, the fraction of the  $38 \mu\text{s}$  dead time per pulse equivalent to  $\mu$  is  $0.17 \mu\text{s}$  and represents the ultimate limit of the pile-up rejection system.

#### 4.2 The decay constant of $^{31}\text{Si}$

The decay constant of  $^{31}\text{Si}$  was determined by processing the  $\gamma$ -spectra recorded during the twelve sequences of counts of the neutron activated  $^{31}\text{Si}$  source. The duration of a single sequence and the number of performed counts,  $N$ , depended on the  $^{31}\text{Si}$  activity at the start of the observation. Specifically, the  $N$  values were 26 (1<sup>st</sup> sequence), 29 (2<sup>nd</sup> sequence), 25 (3<sup>rd</sup> sequence), 28 (4<sup>th</sup>



sequence), 26 (5<sup>th</sup> sequence), 32 (6<sup>th</sup> sequence), 25 (7<sup>th</sup> sequence), 14 (8<sup>th</sup> sequence), 27 (9<sup>th</sup> sequence), 31 (10<sup>th</sup> sequence), 25 (11<sup>th</sup> sequence) and 22 (12<sup>th</sup> sequence).

The relative dead time / input pulse rate varied, on average, from 0.30 / 8300 s<sup>-1</sup> ( $t_c = 460$  s) to 0.06 / 1400 s<sup>-1</sup> ( $t_c = 33$  min) whereas the maximum and the minimum values, 0.31 / 8600 s<sup>-1</sup>, and 0.03 / 550 s<sup>-1</sup>, were reached at the first and last count of the 6<sup>th</sup> sequence which lasted 9.2 h. Furthermore, the ratio of the background count,  $b$ , to the net count,  $n$ , of the <sup>31</sup>Si 1266.1 keV  $\gamma$ -photons,  $\alpha$ , was in the range between 0.010 and 0.035. Since the collected net count values were close to 10<sup>4</sup>, the background counts beneath  $\gamma$ -peak ranged from 100 to 350 (170 in the examples of the 1266.1 keV  $\gamma$ -peaks in figure 2), corresponding to  $\gamma$ -peak background rates from  $9 \times 10^{-4}$  s<sup>-1</sup> to 0.8 s<sup>-1</sup>, respectively.

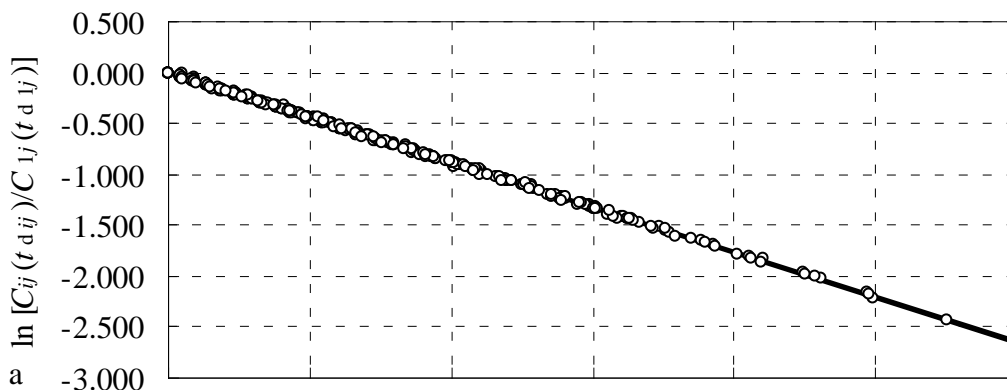
The count rate of the 1266.1 keV  $\gamma$ -photons at the beginning of the  $i^{\text{th}}$  count of the  $j^{\text{th}}$  sequence,  $C_{ij}(t_{dij})$ , is computed according to (1), where  $j = 1, 2, \dots, 12$  and  $i = 1, 2, \dots, N$ . The  $\delta_{ij}$  and  $f_{ij}$  values are obtained from (2).

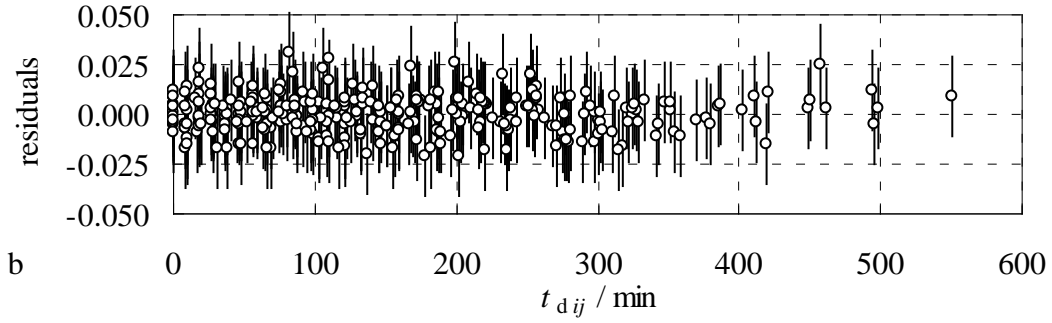
Linear least squares regression is applied to estimate  $\lambda$ , and the corresponding  $t_{1/2}$ , by fitting twelve straight lines to the twelve series of data obtained by taking the natural logarithm of (3) normalized to the  $C_{1j}(t_{d1j})$  value. Specifically, the measured variable,  $y_{ij}$ , is  $\ln[C_{ij}(t_{dij})/C_{1j}(t_{d1j})]$ , the independent variable  $x_{ij}$  is  $t_{dij}$  and the estimated parameters are the twelve intercepts,  $\ln[\hat{C}_j(t_{d1j})/C_{1j}(t_{d1j})]$ , and the shared slope,  $-\lambda$ . Since  $y_{ij}$  depends on  $\lambda$ , the solution is obtained iteratively until convergence.

The main contribution to the uncertainty of  $C_{ij}(t_{dij})$  is the 1% relative uncertainty of the 1266.1 keV  $\gamma$ -peak net count. Therefore, the standard deviation of the error term of  $y_{ij}$ ,  $\varepsilon_{ij} / C_{ij}(t_{dij})$ , is constant and equal to 0.01.

The regression analysis was performed with the Levenberg Marquardt algorithm of the OriginLab<sup>®</sup> software using the line model in global multi-data fit mode [11]. The twelve straight lines fitting the data and the residuals plotted with respect to  $t_{dij} - t_{d1j}$  are shown in figure 5a and figure 5b, respectively. The error bars confirm that the logarithm of uncertainties due to counting statistics are constant.

The resulting <sup>31</sup>Si decay constant,  $\lambda$ , is  $4.4103 \times 10^{-3}$  min<sup>-1</sup> with an associated (fitting) uncertainty of  $0.0046 \times 10^{-3}$  min<sup>-1</sup>. It should be noted that the multi-data fitting of twelve repeated observations of <sup>31</sup>Si lasting about 3 half-lives allowed to average the effect of counting statistics and to reach a 0.1% (fitting) uncertainty.





**Figure 5.** The twelve straight lines (a) and the residuals (b) obtained by fitting the data collected to determine the  $^{31}\text{Si}$  decay constant. The error bars indicate a 95% confidence interval due to counting statistics.

### 4.3. Uncertainty

In the case of an ideal linear relationship between the activity of the source and the counting rate of the detection system, the residuals are expected to be pure stochastic and due to the Poisson nature of radioactive decay. Thus, the only contribution to the uncertainty of  $\lambda$  would be the fitting. However, medium- (cyclic) and low-frequency (drift) deviations must carefully be considered because they can significantly affect the uncertainty also when they are hidden in the residuals. The most important sources of these deviations are (i) instability of the detection efficiency, (ii) pile-up and dead time, (iii) geometrical changes in source positioning, (iv) spectral interference and (v) background signals [2].

The effect of counting statistics can be anticipated using equations (34) and (37) reported in [2] and based on the number,  $N$ , of activity measurements,  $A$ , and associated relative uncertainties,  $u(A)/A$ , repeated at regular time during an observation lasting  $T$ . Although in this study the activity (count rate) measurements, were not equally spaced in time, the resulting relative uncertainty,  $u(\lambda)/\lambda$ , with  $T = 400$  min,  $N = 26$  and  $u(A)/A = 0.01$  is about 0.003 for a single sequence. The corresponding value for the mean of twelve of these sequences, 0.0009, is close to the evaluated (fitting) relative uncertainty, 0.0010.

The results of the stability test didn't show significant additional variability from any cyclic variations or drift of the detection efficiency system other than the expected statistical 0.001 relative standard deviation. Thus, 30% of this standard deviation is conservatively assigned as potential maximum drift of the system during a single sequence and the corresponding 0.0003 is given as a reasonable upper limit for the contribution to  $u(A)/A$ .

The pile-up correction,  $f$ , obtained from (2) with a pile-up factor,  $\mu$ , of  $4.45 \times 10^{-3}$  and a relative dead time,  $t_{\text{dead}}/t_c$ , of 0.30 (at the start count of a sequence) and 0.06 (at the end count of a sequence) is 1.0013 and 1.0003, respectively. Due to the energy dependence of  $\mu$ , a conservative 30% error is assigned to the (corrected) 0.001 relative variation of  $f$  between the start and end counts of a sequence and leads to an average relative uncertainty,  $\langle u(A)/A \rangle$ , i.e. at  $t = 0$  and  $t = 400$  min, of 0.00015 [2]. Since the counting time,  $t_c$ , and the dead time,  $t_{\text{dead}}$ , are known with a negligible uncertainty, the contribution to  $u(A)/A$  of the dead time correction,  $\delta$ , obtained from (2) is negligible as well.

During a single observation, the position of the  $^{31}\text{Si}$  source was fixed with respect to the detector, therefore the effect of geometrical changes in source positioning is absent.

Given the high-purity of the silicon sample used as a source, the 1266.1 keV  $\gamma$ -peak was free from spectral interference. The Compton continuum underlying the  $\gamma$ -peak was approximately flat and due to natural background (see figure 2). To achieve the 0.01 relative uncertainty due to counting statistics, the net count of the  $\gamma$ -peak,  $n$ , reached a constant value (close to  $10^4$ ) in every count. Hence, the effect of a possible and undisclosed departure of the  $\gamma$ -peak from the Gaussian shape is considered constant during the sequence. In addition, the upper limit for the background contamination fraction of the fitted  $\gamma$ -peak area is fixed at 10% of the maximum background to the net count ratio,  $\alpha$ , occurred during the experiments, i.e. 0.0035. The 30% variation of  $\alpha$  usually observed between the start and end of a sequence leads to an average relative uncertainty,  $\langle u(A)/A \rangle$ , i.e. at  $t = 0$  and  $t = 400$  min, of about 0.00053.

In agreement with [2], a propagation factor  $2/\lambda T$  is assumed for the  $u(A)/A$  due to (i) variations or drift of the detection efficiency system, (ii) pile-up correction and (iii) background contamination. The corresponding relative uncertainties,  $u(\lambda)/\lambda$ , are 0.00034, 0.00018 and 0.00060, respectively; these values and the corresponding contributions to the combined uncertainty are listed in table 1.

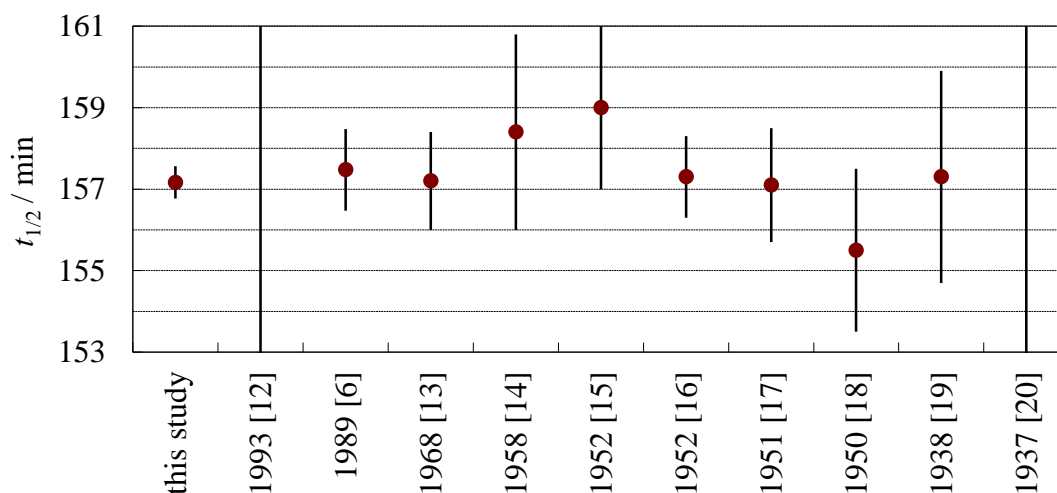
Uncertainty component	$u_i(\lambda)/\lambda$	Index
Counting statistics	0.00104	68%
Instability of the detection system	0.00034	8%
Pile-up and dead time	0.00018	2%
Background signals	0.00060	22%
$u_c(\lambda)/\lambda$	0.00126	100%

**Table 1.** Uncertainty budget for the measurement of the  $^{31}\text{Si}$  decay constant. The resulting combined relative uncertainty,  $u_c(\lambda)/\lambda$ , is also given.

In summary, an overall relative uncertainty of 0.0013 is evaluated for  $\lambda$ . The main contributors are counting statistics, 68%, and background contamination, 22%.

#### 4.4 Comparison with literature data

The presently adopted  $^{31}\text{Si}$  half-life value, 157.36(26) min [5], is the weighted average of ten selected values reported in high quality peer-reviewed journals: 163.0(58) min [12], 157.5(5) min [6], 157.2(6) min [13], 158.4(12) min [14], 159(1) min [15], 157.3(5) min [16], 157.1(7) min [17], 155.5(10) min [18], 157.3(13) min [19] and 170(10) min [20]. These values are shown in figure 6 and compared with the result obtained in this study, i.e. 157.16(20) min.



**Figure 6.** The literature  $^{31}\text{Si}$  half-life values compared with the result obtained in this study. The reference and the publication year are reported. The error bars indicate a 95% confidence interval.

## 5. Conclusions

The  $^{31}\text{Si}$  half-life was determined with repeated measurements of the activity of a  $^{31}\text{Si}$  source activated using neutron irradiation. The value was determined by applying the linear least squares regression of data collected in several decay observations. The possible effects that might affect the result such as instability of the detection system, pile-up, dead-time and background have been limited and, whenever possible, corrected for. The relevant uncertainties were evaluated and propagated following the suggestions reported in [2].

The resulting  $^{31}\text{Si}$  half-life value reached a relative uncertainty of 0.13% and is in agreement with the presently recommended value, given with a 0.16% relative uncertainty, obtained as the weighted average of ten data previously acquired and published with poor information concerning details of experiments and uncertainty budgets.

## Acknowledgements

This work was jointly funded by the Italian ministry of education, university, and research (awarded project P6-2013, implementation of the new SI) and by the European Metrology Research Programme (EMRP) participating countries within the European Association of National Metrology Institutes (EURAMET) and the European Union.

## References

- [1] Karam L, Keightley J and Los Arcos J M 2015 Special Issue Uncertainties in Radionuclide Metrology *Metrologia* **52**
- [2] Pommé S 2015 The uncertainty of the half-life *Metrologia* **52** S51-S65
- [3] De Corte F and Simonits A 2003 Recommended nuclear data for use in the  $k_0$  standardization of neutron activation analysis *At. Data Nucl. Data Tables* **85** 47-67

- [4] D'Agostino G, Di Luzio M, Mana G, Oddone M, Pramann A and Prata M 2015  $^{30}\text{Si}$  Mole Fraction of a Silicon Material Highly Enriched in  $^{28}\text{Si}$  Determined by Instrumental Neutron Activation Analysis *Anal. Chem.* **87** 5716-5722
- [5] Ouellet C and Singh B 2013 Nuclear Data Sheets for  $A = 31$  *Nucl. Data Sheets* **114** 209-396
- [6] Abzouzi A, Antony M S and Ndocko Ndongué V B 1989 Precision measurements of the half-lives of nuclides *J. Radioanal. Chem. Lett.* **135** 1-7
- [7] DSPEC-502<sup>TM</sup> Digital Gamma-Ray Spectrometer *Hardware User's Manual*, Manual Revision D, Advanced Measurement Technology Inc, Berwyn PA 2011
- [8] Gilmore G R 2008 Practical Gamma-ray Spectrometry 2<sup>nd</sup> Edition *Wiley J & Sons* New York 158-159
- [9] *GammaVision*, Version 6.08, Advanced Measurement Technology Inc, Berwyn PA 2006
- [10] Debertin K and Helmer R G 1988 Gamma- and x-ray spectrometry with semiconductor detectors 3<sup>rd</sup> Edition *North-Holland* University of California
- [11] *OriginPro*, Version 9.1.0, OriginLab Corporation, Northampton MA 2013
- [12] McGuire S C, Hossain T Z and Soave R J 1993 1266-keV gamma branch in  $^{31}\text{Si}$  decay *Phys. Rev. C.* **48** 1434-1437
- [13] Reynolds S A, Emery J F and Wyatt E I 1968 Half-Lives of Radionuclides-III *Nucl. Sci. Eng.* **32** 46-48
- [14] Gueben G and Govaerts J 1958 La méthode d'analyse par activation en utilisant les neutrons d'une source Ra-Be *Inst. Interuniv. Sci. Nucléaires*, Bruxelles, Monographie n. **2**
- [15] Motz H T 1952 Beta-Activity of  $^{31}\text{Si}$  and the Masses of the Al and Si Isotopes *Phys. Rev.* **85** 501-502
- [16] De Waard H 1952 A high resolution coincidence instrument with scintillation counters. Delayed and prompt coincidences in the decay of  $^{181}\text{Hf}$  *Phys.* **18** 1151-1156
- [17] Wennerblom A, Zimen K E and Ehn E 1951 Silicon 31: Separation from Irradiated Phosphorus, Half-Life and Beta-Energy *Svensk Kem. Tidskr.* **63** 207-210
- [18] Lüscher E, Ricamo R, Scherrer P and Zünti W 1950 Über die Energieabhängigkeit der Prozesse  $\text{S}^{32}(\text{n p})\text{P}^{32}$  und  $\text{P}^{31}(\text{n p})\text{Si}^{31}$  *Hel. Phys. Acta* **23** 561-565
- [19] Cichochi J and Soltan A 1938 Radiosilicium produit par bombardement du soufre avec des neutrons rapides *Compt. Rend.* **207** 423-425
- [20] Newson H W 1937 The Radioactivity Induced in Silicon and Phosphorus by Deuterium Bombardment *Phys. Rev.* **51** 624-627