Appl. Radiat. Isot. Vol. 44, No. 9, pp. 1209–1211, 1993
Printed in Great Britain. All rights reserved

0969-8043/93 $6.00 + 0.00
Copyright © 1993 Pergamon Press Ltd

Measurement of the Gamma-ray Probability per Decay of Si-31

MARINA F. KOSKINAS and MAURO S. DIAS

Instituto de Pesquisas Energéticas e Nucleares, IPEN, CNEN/SP, Caixa Postal 11049, Pinheiros, CEP 05422-970, São Paulo, Brazil

(Received 26 October 1992; in revised form 19 March 1993)

The gamma-ray emission probability per decay for the 1266-keV transition of ^{31}Si has been measured. This radionuclide was obtained by means of the $^{31}P(n,p)^{31}Si$ reaction in a fast neutron flux at the IPEN 2 MW research reactor. The sample activity has been measured by the $4\pi\beta$-γ coincidence method and the efficiency tracing technique was applied using ^{24}Na as the tracer. The gamma-ray counting has been performed in a calibrated HPGe spectrometer. The resulting emission probability per decay obtained for ^{31}Si was 5.537×10^{-4} with an overall error of 1.21% (one standard deviation).

Introduction

The accurate knowledge of the gamma-ray emission probability per decay (P_γ) of radionuclides is important in several applications. In the case of ^{31}Si, the importance lies mainly in cross section measurements by the activation method (Zijp and Baard, 1979) and in silicon rod monitoring after neutron transmutation doping (Parma and Hart, 1982).

The decay scheme of ^{31}Si is shown in Fig. 1. This nuclide decays 99.94% by beta-transition to the ^{31}P ground state. The remaining 0.06% feeds the first excited state, giving rise to a 1266 keV gamma-ray.

In the literature, the measurements of P_γ for the 1266 keV gamma-ray of ^{31}Si are scarce and show large discrepancies. The main reported values for this parameter are 7×10^{-4} with no stated uncertainty (Lyon and Manning, 1954) and $(5.90 \pm 0.29) \times 10^{-4}$ (Parma and Hart, 1982). For this reason we have undertaken the measurement of this emission probability using $4\pi\beta$-γ coincidence counting and HPGe spectrometric techniques. Because of the very low intensity of the 1266 keV gamma-ray, direct use of the $4\pi\beta$-γ coincidence counting is not possible. Therefore, the efficiency tracing technique (Merritt and Gibson, 1978) should be used. This technique has been applied in the present work choosing ^{24}Na as tracer due to its very close beta energy as compared to ^{31}Si. We have reported a preliminary result of $(5.87 \pm 0.14) \times 10^{-4}$ (Geraldo *et al.*, 1992). This measurement has been revised thoroughly and changed slightly. The new value is given in the present paper. The corresponding change in the reported cross section is briefly discussed.

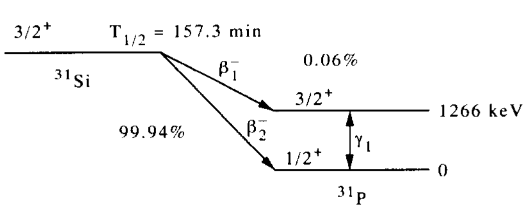

Fig. 1. Decay scheme of ^{31}Si (Zijp and Baard, 1979).

Experimental

Source preparation

The ^{31}Si was obtained by means of the $^{31}P(n,p)^{31}Si$ reaction in a fast neutron flux of $4 \times 10^{12}\, cm^{-2}\, s^{-1}$ obtained near the core of the IPEN 2 MW research reactor. The sample was made of 150 mg NH_4PO_4 powder wrapped in aluminum sheet and sealed inside a polyethylene tube. A cadmium cover was used around the tube in order to avoid undesirable activation by thermal neutrons. The irradiation time was around 1 h. After irradiation, the NH_4PO_4 powder was measured in a HPGe spectrometer system. Immediately after this, the powder was dissolved into an accurately known mass of 1 M HCl solution.

The ^{31}Si sources to be measured in the $4\pi\beta$-γ system were prepared by dropping known aliquots of the solution on a $20\,\mu g/cm^2$ thick COLLODION film. This film had been previously coated with a $10\,\mu g/cm^2$ gold layer in order to turn the film conductive. A seeding agent (CYASTAT SM) was used to improve the deposit uniformity and the sources were dried in a warm (40°C) nitrogen jet (Wyllie *et al.*, 1970). The accurate source mass determination was performed using the picnometer technique (Campion, 1975). The tracer solution of ^{24}Na was prepared by dissolving $NaCO_2$ in a 1 M HCl solution after irradiation with thermal neutrons. The ^{24}Na sources were prepared in the same way as for ^{31}Si.

For the first irradiation, three sets of six sources each were prepared: one from the ^{31}Si solution, one from the ^{24}Na solution and another one mixing drops from both solutions (mixed sources). For the remaining irradiations, only ^{31}Si sources were prepared.

4$\pi\beta$-γ Measurements

The system used for absolute standardization of the ^{31}Si sources consists of a gas-flow proportional counter with 4π geometry (beta-channel) coupled to a pair of NaI(Tl) scintillation counters (gamma-channel) operating in coincidence (Moura, 1969). The proportional counter plateau was reached with a +2050 V bias. The lower discrimination level was set around 0.9 keV. The gamma-channel window was set to measure only the 1369-keV total absorption peak of ^{24}Na.

The presence of ^{32}P from (n,γ) reaction in ^{31}Si and mixed sources has been taken into account by remeasuring these sources several days after irradiation. The amount of ^{32}P was then calculated and subtracted from the observed ^{31}Si counting rate in the $4\pi\beta$ detector. The magnitude of the ^{32}P beta contribution at the time of measurement was around 6.5%. The contribution from ^{32}Si produced by (n,γ) reaction in ^{31}Si has been estimated to be <0.01%.

The ^{31}Si specific activity (Bq/g) measured by a mixed source was deduced applying the equations given in (Merrit and Gibson, 1978) and resulted:

$$A_{31} = N_{\beta 31}/(\epsilon_\beta \cdot m_{31}) \qquad (1)$$

where:

$$N_{\beta 31} = N_{\beta \mathrm{TOT}} - A_{24} \cdot \epsilon_\beta \cdot m_{24} - N_{\beta 32} \qquad (2)$$

and $N_{\beta \mathrm{TOT}}$ is the total counting rate observed in the 4π proportional counter, corrected for dead time and background, A_{24} is the ^{24}Na specific activity determined by the $4\pi\beta$-γ coincidence method, corrected for decay, ϵ_β is the β efficiency calculated by the N_c/N_γ ratio for the given mixed source, where N_c/N_γ is the ratio of coincidence and gamma rates, calculated by the Cox–Isham (Cox and Isham, 1977) formalism, $N_{\beta 32}$ is the counting rate due to ^{32}P, corrected for dead time, background and decay, and m_{31} and m_{24} are the masses of the ^{31}Si and ^{24}Na aliquots contained in the mixed source, respectively.

Figure 2 shows the behavior of the observed specific activity of ^{31}Si for several mixed sources, as a function of the inefficiency parameter $(1 - N_c/N_\gamma)/(N_c/N_\gamma)$. It can be seen that N_c/N_γ is very close to unity for all sources. A statistical analysis showed that the calculated slope is zero within the measured errors. For this reason, for the remaining three irradiations only ^{31}Si sources were measured. In this case the specific activity is given by:

$$A_{31} = (n_{\beta \mathrm{TOT}} - N_{\beta 32})/(\bar{\epsilon}_\beta \cdot m_{31}) \qquad (3)$$

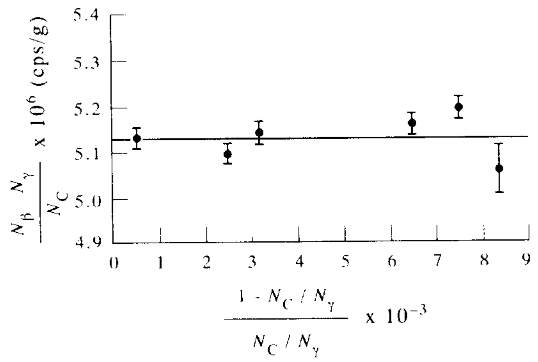

Fig. 2. Behavior of the observed specific activity of mixed sources as a function of the inefficiency parameter: $(1 - N_c/N_\gamma)/(N_c/N_\gamma)$.

where: $\bar{\epsilon}_\beta$ is the average β efficiency determined by combining the N_c/N_γ ratios of all mixed sources. Its value is (0.995 ± 0.003).

The activity of the irradiated sample was calculated by taking the specific activity multiplicated by the total mass of the ^{31}Si solution (50 g), which was measured with an error of about 0.1%.

Gamma Spectrometry Measurements

The ^{31}Si sources were measured in HPGe spectrometer. The detector volume was 99.7 cm³ and the intrinsic efficiency was 20%. This spectrometer has been calibrated in the energy range between 778.90 and 1408.02 keV by measuring IAEA standard sources of ^{60}Co and ^{152}Eu. The calibration was checked by measuring a ^{60}Co source standardized in the $4\pi\beta$-γ coincidence system.

The source to detector distance was around 17.2 cm. At this distance the cascade-summing effects were estimated to be negligible. Dead time and pile-up corrections were applied by measuring a reference pulser peak near the upper end of the gamma spectrum simultaneously with the sources. The area of the peak was evaluated by summing all MCA counts in the interval $[C - 3\delta, C + 3\delta]$ where C is the peak centroid and δ is the FWHM (full width

Table 1. Estimated uncertainties envolved in the ^{31}Si γ-ray emission probability measurement (in %)

Source of error	Irradiation 1	2	3
Peak area statistics	0.84	0.99	0.85
Source mass	0.15	0.15	0.15
Attenuation factor	0.15	0.15	0.15
Geometric factor	0.52	0.52	0.52
$N_{\beta 31}$	0.15	0.22	0.80
Half-life	0.40	0.23	0.11
$N_{\beta 32}$	0.17	0.17	0.38
Decay time	0.44	0.44	0.44
β efficiency	0.30	0.30	0.30
γ efficiency	0.54	0.54	0.54
Total error	1.60	1.65	1.79

Table 2. Results of the gamma-ray emission probability of ^{31}Si

Irradiation	I_γ ($\times 10^{-4}$)	Error (%)
1	5.532	1.60
2	5.473	1.65
3	5.632	1.79
Average	5.537	1.21

at half maximum). The net counts were obtained by considering the background in a region of width δ at both sides of the gamma-ray peak. Corrections were applied to account for geometric differences between the ^{31}Si and the standard sources as well as for gamma-ray self absorption.

Results and Discussion

Table 1 shows the estimated uncertainties involved in the measurement of P_γ for the 1266-keV transition of ^{31}Si. As it can be seen, the main sources of uncertainty are in the HPGe measurement, namely: statistics in the 1266 MeV γ-ray peak area, detector efficiency and geometric corrections. The overall error for each irradiation was in the range of 1.6–1.8%. Table 2 shows the average value for P_γ of ^{31}Si. This value was calculated by a least square fit procedure, applying the covariance methodology. The result was $(5.537 \pm 0.067) \times 10^{-4}$. The reduced chi-square was 1.82 indicating a satisfactory fit. The difference between this value and the preliminary value $(5.87 \pm 0.14) \times 10^{-4}$ reported previously (Geraldo *et al.*, 1992) comes from a more rigorous account of the ^{32}P activity and improvements in the gamma-spectrometric measurements. The result of the present work is somewhat smaller than reported previously by Parma and Hart (1982) and Lyon and Manning (1954).

With this new value of P_γ, the reported cross section for the $^{31}P(n,p)^{31}Si$ reaction (Geraldo *et al.*, 1992) is changed to (36.0 ± 1.4) mb. This value is in better agreement with the results of Fabry *et al.* (1978) and Callammand (1974).

Acknowledgements—The authors would like to thank Dr Luiz P. Geraldo for the valuable discussions on covariance analysis, to Ms Katia A. Fonseca and Ms Nora L. Maidana for the participation in the measurements and to Ms Eliane Pocobi for the careful sample preparations.

References

Callamand A. (1974) Cross sections for fission neutron induced reactions. In *Handbook on Nuclear Activation Cross Sections*, p. 273. Technical Report Series No. 156, IAEA, Vienna.

Campion P. J. (1975) *Procedures for accurately diluting and dispensing radioactive solutions*, Monographie BIPM-1, Bureau International des Poids et Mesures, Sevres.

Cox D. R. and Isham V. (1977) A bivariate point process connected with electronic counters. *Proc. Roy. Soc.* **A356**, 149.

Fabry A., McElroy W. N., Kellog, L. S., Lippincott E. P., Grudl J. A., Gilliam D. M. and Hansen G. E. (1978) Review of Microscopic Integral Cross Section Data. In *Fundamental Neutron Cross Sections for Reactor Dosimetry* Vol. 1, p. 233. Review Papers, IAEA-208, Vienna.

Geraldo L. P., Dias M. S. and Koskinas M. F. (1992) Average neutron cross section measurements in U-235 fission spectrum for some threshold reactions. *Radiochim. Acta* **57**, 63.

Lyon W. G. and Manning J. J. (1954) *Phys. Rev.* **93**(3).

Merritt J. S. and Gibson F. H. (1978) Standardization of ^{137}Cs by the $4\pi(PC)\beta$-γ efficiency-tracing method with ^{134}Cs as tracer. Report AECL-6203, Atomic Energy Canada Limited.

Moura L. P. (1969) Generalized Coincidence Method for Radionuclide Absolute Activity Measurement. Ph.D. Thesis, University of Campinas (in Portuguese).

Parma E. J. Jr and Hart R. R. (1982) Measurement of the Gamma Abundance of Silicon-31. In *Proc. 4th Neutron Transmutation Doping Conference*, p. 127. NBS. Gaithersburg.

Wyllie H. A., Johnsons E. P. and Lowenthal G. C. (1970) *Int. J. Appl. Radiat. Isot.* **21**, 490.

Zijp W. L. and Baard J. H. (1979) Nuclear Data Guide for Reactor Neutron Metrology. In Commission of the European Communities, *Nuclear Science and Technology* (EUR-7164 EN Part I and II) Brussels.