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Methodology for monitoring the residual activity in silicon rods irradiated with thermal neutrons

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Abstract

A calibrated detection system developed for monitoring the residual activity in silicon rods irradiated for neutron transmutation doping is described. This residual activity is mainly due to ^{31}Si and ^{32}P, produced by thermal neutron capture reactions. The set-up consists of a scanning device, developed for positioning the rods, coupled to a GM detector for measuring β-rays coming from the rod. The set-up has been calibrated using a ^{32}P aqueous solution standardised in a $4\pi\beta$ gas-flow proportional counter system. The validity of the experimental procedure has been checked by Monte Carlo calculations. An additional check has been made by measuring the residual activity of a silicon rod irradiated in a well-known neutron fluence and the formalism used for calculating the activity on the basis of irradiation parameters is given. The results are compared to the radiological limit imposed by IAEA regulations in order to consider the material as exempt before it could be released for industrial applications.

1. Introduction

During the neutron transmutation doping (NTD) process in silicon rods, the radionuclides ^{31}Si and ^{32}P are produced by neutron capture reactions. The residual activity of these radionuclides must fall below radiological limits, before the rods can be released for industrial applications.

Silicon-31 is a $\beta-\gamma$ emitter with a half-life of 157.3 min and a maximum β energy of 1492.02 keV (Firestone and Shirley, 1996). This radionuclide emits a 1266.12 keV γ-ray of very low intensity (0.07%) (Firestone and Shirley, 1996) making it difficult to be detected. Phosphorus-32 is a pure β emitter, with a half-life of 14.262 d and a maximum β energy of 1710.6 keV (Firestone and Shirley, 1996). The maximum range of these β particles in silicon is 3.8 mm or less (Knoll, 1989), therefore only β-rays emitted near the rod surface can be detected. The resulting counting rate must be calibrated against the specific activity of ^{31}Si or ^{32}P inside the silicon rod.

In the present work, the experimental calibration of a detection system designed for monitoring the induced activity of these radionuclides in irradiated silicon rods is described. The experimental procedure has been checked by Monte Carlo calculation. An additional check has been made by measuring the residual activity of a silicon rod irradiated in a well-known neutron fluence.

2. The irradiation process

A detailed description of the irradiation rig developed for NTD at the IEA-R1 research reactor has been published elsewhere (Carbonari et al., 1993). Crystal ingots with 3 and 4 in diameter and 40 cm long are irradiated typically for 20–30 h in a thermal neutron flux of the order of $10^{13}\,\mathrm{n\,cm^{-2}\,s^{-1}}$.

The radionuclides ^{31}Si and ^{32}P are produced by the following process:

$$^{30}\mathrm{Si(n,\gamma)}^{31}\mathrm{Si} \xrightarrow{\beta^-} 31\mathrm{P(n,\gamma)}^{32}\mathrm{P} \xrightarrow{\beta^-} 32\mathrm{S}.$$

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The main reaction and decay parameters are listed in Table 1. The residual activity from other reactions has been estimated to be negligible (Sebastião, 1992).

The activity of ^{31}Si after an irradiation time t in a thermal neutron flux Φ is given by

$$A_1 = \lambda_1 N_0 \sigma_0 \phi (1 - e^{-\lambda_1 t}), \quad (1)$$

where N_0 is the atomic density of ^{30}Si in the rod, σ_0 is the averaged thermal neutron capture cross sections of ^{30}Si, and λ_l is the decay constant of ^{31}Si.

For the same irradiation conditions, the activity of ^{32}P is given by

$$A_2 = \lambda_2 N_0 \sigma_0 \sigma_1 \phi^2 \left\{ \frac{1 - e^{-\lambda_2 t}}{\lambda_2 \sigma_1 \phi} - \frac{e^{-\sigma_1 \phi t} - e^{-\lambda_2 t}}{\sigma_1 \phi (\lambda_2 - \sigma_1 \phi)} + \frac{e^{-\lambda_1 t} - e^{-\lambda_2 t}}{(\lambda_2 - \lambda_1)(\lambda_1 - \sigma_1 \phi)} - \frac{e^{-\sigma_1 \phi t} - e^{-\lambda_2 t}}{(\lambda_1 - \sigma_1 \phi)(\lambda_2 - \sigma_1 \phi)} \right\}, \quad (2)$$

where σ_1 is the averaged thermal neutron capture cross sections of ^{31}P, and λ_2 is the decay constant of ^{32}P.

If the irradiation time is much shorter than the ^{32}P half-life, then Eq. (2) can be simplified and becomes:

$$A_2 = \tfrac{1}{2} \lambda_2 N_0 \sigma_0 \sigma_1 \phi^2 t^2. \quad (3)$$

At the end of an irradiation time of 30 h in a thermal flux of 1.0×10^{13} n cm^{-2} s^{-1}, the calculated ^{32}P activity (A_2) results 4.1 Bq g^{-1}. This value is smaller than the radiological limit of 1 kBq g^{-1} imposed by IAEA regulations (IAEA, 1994) in order to classify the material as exempt. Therefore, ^{32}P does not represent a problem, except when the rod has appreciable ^{31}P content before irradiation. In this case the additional activity can be calculated by Eq. (1), using the corresponding parameters for the ^{31}P(n, γ) ^{32}P reaction. On the other hand, the amount of ^{31}Si activity after the same irradiation time is about 52 kBq g^{-1}. This activity should be allowed to decay, and has to be checked before releasing the rods for industrial applications.

3. Experimental calibration

A scanning device shown in Fig. 1 has been developed for silicon rod monitoring. The rod is positioned horizontally and can move along the three co-ordinates: x, y and z. It can also rotate around the z-axis in order to scan the curved surface and other end of rod.

The monitor was a Geiger–Müeller counter having 5.3 cm^2 sensitive area and a 1 μm-thick mica entrance window. The detector was also placed horizontally in a fixed position with respect to the scanning device. A cylindrical collimator made of Lucite with 2.5 cm internal diameter and 1.2 cm long was positioned in front of the detector window, in order to have a well defined source–detector geometry.

The ^{31}Si β-ray energy is only about 15% lower as compared to ^{32}P. Because the calibration using ^{32}P is much easier to achieve due to the longer half-life and simple experimental procedure, this radionuclide has been used for calibrating the system. Corrections were calculated by Monte Carlo, in order to obtain the calibration factor for ^{31}Si.

A ^{32}P solution was standardised by means of a $4\pi\beta$ gas-flow proportional counter. The radioactive sources were prepared by depositing known aliquots of the solution on Collodion substrate 10 μg cm^{-2} thick. Corrections were applied to account for background, decay, dead time and self-absorption. The standard ^{32}P radioactive solution was placed in a cylindrical container made of PVC, 7.62 cm in diameter and 25 cm long. This container had two windows 3.1 cm in diameter and 8 μm thick made of Makrofol KG located at the flat end and curved surface of the container walls. The container was flush to the detection system and the window was coaxial to the detector collimator (see Fig. 1).

In this geometry, the thickness of water can be considered infinite in comparison to the β range.

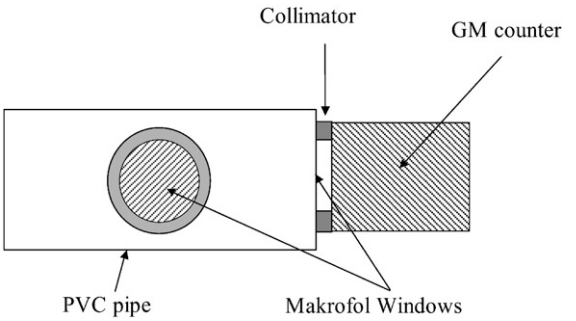

Fig. 1. Schematic diagram of the experimental set-up for monitoring irradiated silicon rods.

Table 1
Nuclear reaction cross sections and decay parameters involved in the production of ^{31}Si and ^{32}P

Reaction	Thermal neutron cross section (IAEA, 1987)	Decay product half-life (Firestone and Shirley, 1996)
^{30}Si (n,γ) ^{31}Si	0.107 ± 0.002	157.3 ± 0.3 min
^{31}P (n,γ) ^{32}P	0.172 ± 0.006	14.262 ± 0.014 d

Therefore, since the source–detector distance and the collimator are fixed, the GM counting rate is proportional to the specific activity (in $\mathrm{Bq\,g^{-1}}$) and the experimental detection efficiency can be given by

$$\varepsilon_{\mathrm{exp}} = \frac{C}{A}, \tag{4}$$

where C is the GM detector counting rate (cps) corrected for background, and A is the specific activity of ^{32}P standard solution ($\mathrm{Bq\,g^{-1}}$).

This efficiency is, in principle, applicable only to water as absorption medium. An independent determination must be performed in order to verify possible differences if silicon is used as the absorption medium. This has been done by Monte Carlo calculations as described in the following section.

4. Monte Carlo calculations

The electron trajectory inside a solid or liquid medium is very complex. Elastic, inelastic scattering and absorption processes may take place, resulting in a large number of interactions inside the medium. In the present work, a simple approach has been developed using a Monte Carlo technique, in order to estimate the detection efficiency for the case of water (and silicon) as the absorption medium.

In the calculation, several steps have been followed. First, a random point of emission has been chosen inside the cylindrical source within the maximum β-particle range. The distance from the source surface to the emission point in the axial direction is given by

$$z = \xi \;\; R, \tag{5}$$

where ξ is an uniform random number in the]0,1[interval, and R is the maximum β-particle range given by (Price, 1964)

$$R = 0.407E^{1.38}, \tag{6}$$

where E is the characteristic β energy in MeV.

The next step is to select a direction for the β-ray. The direction cosines in the x, y and z directions are respectively given by

$$u = \rho\cos\phi,$$

$$v = \rho\sin\phi$$

and

$$w = 1 + (w_{\max} - 1)\xi, \tag{7}$$

where

$$w_{\max} = \cos\{\tan^{-1}[(r_{\mathrm{s}} + r_{\mathrm{d}})/d]\}.$$

$r_{\mathrm{s}}, r_{\mathrm{d}}$ and d are the source radius, detector radius and source–detector distance, respectively;

$$\rho = \sqrt{1 - w^2},$$

$$\phi = \pi \;\; (2\xi - 1)$$

and ξ are random numbers in the]0,1[interval.

The procedure for obtaining w described above is used in order to improve the detection efficiency. A correction factor for the solid angle ($\Omega_{\max}$) is applied as shown in Eq. (10). If the direction taken by the β particle is within the solid angle subtended by the detector, the event is accepted and a transmission factor is calculated by

$$f = \exp(-\mu x), \tag{8}$$

where x is the distance travelled by the β-ray from the source to the detector, and μ is the β-ray attenuation coefficient given by (Evans, 1969)

$$\mu = \frac{a}{E^b}, \tag{9}$$

where E is the maximum β-ray energy (in MeV) and a, b are constants taken from the literature (Baltakmens, 1970).

This simulation does not consider individual β-rays. Each emission in the source is treated as a β-ray beam and the output is the average transmission through the path between source and detector. All materials along the path were taken into account and the μ value (in $\mathrm{cm^2\,g^{-1}}$) has been approximated in the same manner as for aluminium.

The theoretical detection efficiency $\varepsilon_{\mathrm{th}}$ ($\mathrm{cps\,g\,Bq^{-1}}$) is given by

$$\varepsilon_{\mathrm{th}} = \Omega_{\max}\frac{1}{n}\sum_{i=1}^{m} f_i, \tag{10}$$

where $\Omega_{\max} = \frac{1}{2}(1 - w_{\max})$, n is the number of histories followed, f_i is the transmission factor for each accepted history, and m is the number of accepted histories.

5. Results and discussion

The measured specific activity of the ^{32}P solution measured in the $4\pi\beta$ counter was $6.89\,\mathrm{kBq\,g^{-1}}$, with an overall uncertainty of 5% (at 68% confidence level). A series of 10 measurements has been performed in order to obtain the GM counting rate corresponding to the standardised ^{32}P solution. The average value was 138.9 ± 0.7 cps. From Eq. (4), one can calculate the experimental detection efficiency of the GM calibration system, the resulting value is shown in Table 2.

For estimating μ from Eq. (9), the values of a and b were 16.5 and 1.40, respectively (Baltakmens, 1970). Table 2 shows the results of the theoretical detection efficiency for water and silicon, according to Eq. (10). The calculated values agree well with each other within the estimated overall uncertainty in the calculation (9.7%). The detection efficiency for ^{31}Si is 35% lower than the corresponding value for ^{32}P, as shown in Table 2.

Table 2
Detection efficiencies of GM detection system

Radionuclide	Absorption medium	Detection efficiency $\times 10^{-2}$ (cps g Bq^{-1})
^{32}P	Water (experimental)	2.079 ± 0.052
	Water (theoretical)	2.243 ± 0.005
	Silicon (theoretical)	2.213 ± 0.005
^{31}Si	Silicon (theoretical)	1.422 ± 0.004

The indicated uncertainties in the calculation are statistical only (one standard deviation). The overall uncertainty in the theoretical efficiency was estimated to be 9.7%.

Despite the simplicity of the theoretical model chosen, the agreement between the calculated and the experimental results for water is good. Therefore, no correction factor has been applied to the experimental efficiency.

The main contribution to the absolute uncertainty in the theory comes from the β attenuation coefficient (7.2%), estimated by fitting the values for aluminium in the literature. Nevertheless, since the procedure used to calculate the efficiency is the same for both absorption media, the relative uncertainty is expected to be lower.

An additional check in the calibration procedure has been performed by irradiating a silicon rod in a well-known neutron fluence. The rod was positioned in a region near the IEA-R1 research reactor core where the neutron flux had been measured accurately by the activation method, using the ^{59}Co(n,γ) ^{60}Co reaction (Carbonari et al., 1993). After decaying several days, the silicon rod was monitored by a large GM detector (pancake-type) positioned close (3 mm) to the detector window, by means of a Lucite collimator 44 mm in diameter. The resulting net counting rate was 1.76 ± 0.24 cps, corrected for background and decay. The Monte Carlo calculated efficiency for this geometry was 0.33 ± 0.06 cps g Bq^{-1}. The large uncertainty in this value is due to the presence of a metal grid in front of the GM window, giving rise to β-ray scattering and attenuation effects which are difficult to estimate. The resulting specific activity was 5.3 ± 1.3 Bq g^{-1}. This value agrees well with the one calculated by means of Eq. (3) which yielded 4.5 ± 0.4 Bq g^{-1}.

For the set-up shown in Fig. 1, the room background in a 1 min measuring time resulted in a counting rate of 1.50 ± 0.08 cps. The detection limit taken as three times the standard deviation of this value yields 0.24 cps, corresponding to a minimum detectable specific activity of 12.3 Bq g^{-1} for ^{32}P and 14.9 Bq g^{-1} for ^{31}Si. These values are much lower than the radiological limit of 10^3 Bq g^{-1} imposed by IAEA regulations, showing that the proposed set-up is adequate for monitoring purposes. Nevertheless, improved sensitivity may be achieved by detector shielding, a larger detector window or a shorter distance between rod and detector. Variations in the detection geometry can easily be incorporated in the calculations, by changing input parameters in the code.

Surface contaminations produced by neutron capture reactions are usually from β-emitters and can be also checked using the present set-up. In this case, the calculated detection efficiency is smaller than expected experimentally because there is much less β-ray self-absorption. The present result, therefore, gives a conservative upper limit for the residual activity present on the rod surface.

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References

Baltakmens, T. 1970. A simple method for determining the maximum energy of beta emittes by absorption measurements. Nucl. Instrum. Methods 82, p. 264.

Carbonari, A.W., Pendl Jr., W., Sebastião, J.R., Saxena, R.N., Dias, M.S., 1993. An irradiation rig for neutron transmutation doping of silicon in the IEA-R1 research reactor. Nucl. Instrum. Methods B 83, 157.

Evans, R.D., 1969. The Atomic Nucleus, 12th Edition. McGraw-Hill, New York.

Firestone, R.B., Shirley, V.S., 1996. Table of Isotopes, CD ROM edition.

IAEA, 1987. Handbook on Nuclear Activation Data. IAEA Technical Reports Series No. 273, Vienna, p. 208.

IAEA, 1994. International Basic Safety Standards for Ionising Radiation and for the Safety of Radiation Sources, Safety Series No. 115-I, Vienna, 1994, p. 84.

Knoll, G.F., 1989. Radiation Detection and Measurement, 2nd Edition. Wiley, New York.

Sebastião, J.R., 1992. M.Sc. Thesis, IPEN, São Paulo, 1992.

Price, W.J., 1964. Nuclear Radiation Detection, 8th Edition. McGraw-Hill, New York, p. 19.