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An irradiation rig for neutron transmutation doping of silicon in the IEA-R1 research reactor

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An irradiation rig with simple design has been constructed and installed in the IEA-R1 research reactor for neutron transmutation doping (NTD) of silicon with phosphorus. Crystal ingots with 3 and 4 in. diameter can be irradiated in this device. By adopting a procedure in which two ingots of 20 cm long each are irradiated simultaneously and their positions interchanged at a point when precisely half the total necessary neutron dose has been received, it has been possible to achieve the desired axial uniformity of the neutron dose. This method avoids the use of neutron absorbing shields around the crystals which necessarily compromise the overall irradiation capacity of the reactor. Test irradiations were performed with 50 float zone silicon crystals, and the results of radial and axial uniformities in the final resistivity values as well as the doping accuracy obtained in the test irradiations show an excellent doping quality achieved.

1. Introduction

Pure silicon is the most commonly used material for the fabrication of electrical and electronic devices, ranging from large volume high voltage thyristors, power diodes and transistors to integrated circuits for computers and microprocessors. However, in order to be used in these devices, the silicon needs to be doped with another element to achieve the desired resistivity value which usually differs for different applications. The most commonly used dopants are phosphorus (for n-type silicon) and boron (for p-type silicon). The conventional phosphorus doping is carried out by the incorporation of the dopant in the required concentration in the molten stage during crystal growth. The conventional doping however leads to inhomogeneous distribution of the dopant due to the low distribution coefficient of phosphorus in silicon, and the resultant material presents large resistivity variations. As a consequence this material is unsuitable for the production of high volume devices such as rectifiers for high-voltage direct current (HVDC) transmission due to the hot spot formation and eventual possibility of device breakdown.

The neutron transmutation doping (NTD) method can produce silicon semiconductors with perfectly uniform dopant distribution through irradiation of crystal with thermal neutrons in a nuclear reactor. The NTD process is based on the capture of thermal neutrons by the ^{30}Si nuclei (3.1% in natural silicon) producing the radioactive nuclei ^{31}Si which decay with a half-life of 2.62 h to the stable isotope ^{31}P,

$$^{30}Si(n, \gamma)^{31}Si \rightarrow {}^{31}P + \beta^{-}. \quad (1)$$

As other possible nuclear reactions contribute to a negligible extent, the transmutation nuclear reaction produces n-type semiconductor silicon. Moreover, since the irradiation of a silicon crystal can be performed in an homogeneous way in nuclear reactors, the resistivity variation is very small throughout the volume of the crystal.

The possibility of doping silicon with phosphorus by neutron irradiation was first pointed out by Lark-Horovitz in 1951 [1]. Ten years later, Tanenbaum and Mills [2] carried out the first neutron transmutation doping experiment irradiating small pieces of silicon to study the phosphorus distribution. In the beginning of the seventies, NTD float zone silicon was used for the fabrication of the first high power devices followed by extensive research which showed the superiority of the resulting device characteristcs [3–5]. Since then, neutron transmutation doping has been offered by a number of research reactors, mainly in Europe and the USA. The total world production of NTD silicon is estimated to be over 100 tons annually [6].

With the increasing demand for NTD silicon with very tight target resistivity tolerances, and considering

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the fact that only a limited number of research reactors are able to irradiate silicon presently, it is important to improve the silicon irradiation process by introducing new procedures and precise neutron dose control methods. The present work describes a very efficient and accurate method of irradiating silicon crystals without the use of neutron absorbing shields or moving the crystals in the axial direction. Radial and axial uniformities, doping accuracy and the simplicity of operation are the main objectives which were considered in the design of a test silicon irradiation facility at the IEA-R1 reactor.

2. The IEA-R1 research reactor

The IEA-R1 reactor is an open swimming-pool-type light water reactor operated at 2 MW on an 8 h day, 5 days per week, cycle. The reactor core is comprised of 31 MTR-type fuel elements including four elements with neutron absorbing control plates. About half of the fuel elements are 93% ^{235}U while others use 20% enriched uranium. The reactor core is surrounded by graphite blocks serving as a reflector. A cross-sectional view of the reactor is shown in fig. 1. The silicon irradiation position is on the top left side.

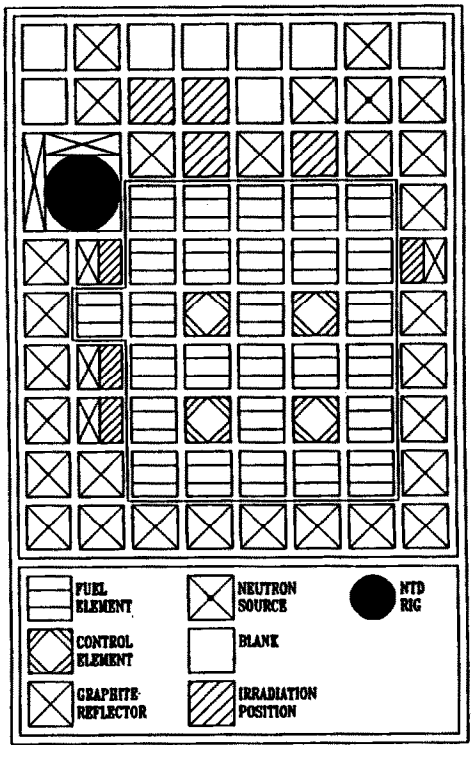

Fig. 1. A cross-sectional view of the IEA-R1 research reactor core.

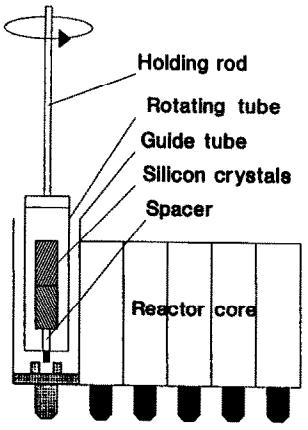

Fig. 2. A side view of the silicon irradiation rig located in the graphite reflector.

3. Silicon irradiation rig

Fig. 2 is a side view of the silicon irradiation rig which is located in the graphite reflector. The rig essentially consists of two parts, the one fixed on the reactor grid plate is a square cross-sectional aluminium guide tube with dimensions of 12 cm × 12 cm × 150 cm. The other part of the rig consists of a freely rotating cylindrical aluminium tube which is 175 cm long, has 109 mm internal diameter and a 2.5 mm thick wall. The cylindrical tube is supported inside the guide tube through an aluminium pin which holds it at the centre during rotation. The rotating tube is connected to a long aluminium rod which extends above the pool water, and is coupled to the shaft of an electric motor through a ball bearing. The rotating cylinder serves as the irradiation tube for silicon crystals. The rotation speed is 2 rpm, and to ensure that the tube does in fact rotate during the irradiation period the rotational motion is constantly monitored through an optical sensor.

All the silicon crystals are encapsulated in aluminium cans which are 50 cm long and have 0.5 mm wall thicknes. The internal diameter of the can varies according to the diameter of the silicon crystal to be irradiated (3 or 4 in.). The aluminium cans are provided with holes at the bottom and at the top to permit the flow of water around the crystal during irradiation.

The neutron dose is controlled by two silver wire self-powered neutron detectors (SPND) placed very close to the irradiation tube. The neutron detectors utilize the β-decay of the radioactive silver isotopes $^{108,110}Ag$ to produce a current which is proportional to

the neutron flux. The current is measured by a digital current integrator and counted in a preset scaler. When the integrated counts reach the preset value, an alarm sounds prompting the reactor operator to take the necessary action to pull the irradiation tube out of the reactor core. The self-powered neutron detectors are calibrated periodically against cobalt wire monitors irradiated simultaneously. The activity of the cobalt monitors is measured by an ionization chamber.

4. Axial profile of the neutron flux

To obtain the desired resistivity uniformity, the neutron flux must be as homogeneous as possible over the entire crystal volume. While the small radial flux gradient of the reactor can be greatly reduced by rotating the crystal around its axis during the irradiation, which usually results in a radial resistivity variation of the order of 1% or less, the detailed knowledge of the neutron flux variation in the axial direction is essential. The axial flux profile depends on the reactor type and dictates the method to be used for optimizing the maximum to minimum resistivity varaition over the ingot length and consequently the maximum ingot length which can be irradiated.

The thermal neutron flux profile has been measured by the activation method. The selected reaction was $^{59}Co(n, \gamma)^{60}Co$, due to the fact that it is almost insensitive to epithermal neutrons and provides an adequate activity for quick measurement by an ionization chamber or HPGe spectrometer.

Cobalt wire monitors wrapped in thin aluminium sheets were positioned at regular distances on the surface of aluminium rods having the same dimensions as the silicon ingots. Aluminium was chosen because of its similarity to silicon with respect to the neutron scattering and absorption cross-sections. Irradiation of the aluminium rods was carried out by following the same procedure as adopted for the silicon crystals, and the data were obtained for cobalt monitors covered with and without cadmium sheets. The neutron fluence was calculated using the Westcott formalism [7]. Most of the correction factors involved cancelled out, and the only remaining important factor was the cobalt wire self-shielding to neutrons, which was determined in a separate experiment [8] for the used wire. The measured flux profile is shown in fig. 3. The cadmium ratio was determined to be 29.3 ± 0.6.

5. Irradiation method

An inspection of the axial neutron flux profile at the silicon irradiation position (fig. 3) shows that the flux peaking occurs at about the geometrical centre of the

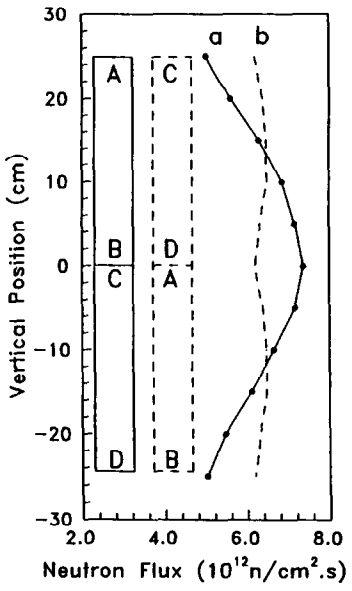

Fig. 3. Experimentally measured thermal neutron flux profile in the vertical direction (a). The final flux profile as would be seen by the crystal pair when their positions are interchanged at 50% of the required dose.

irradiation tube and that the neutron flux drops rather steeply but quite symmetrically above and below this central position reaching about 50% of the peak value at approximately 25 cm on either side.

A decision to utilize the maximum possible irradiation capacity of the rig as well as a consideration of the cost for NTD irradiation set the goal to irradiate at least 40 cm long silicon crystals in the installed facility. The approach to axial uniformity involving flux profile modification using neutron absorbers was immediately rejected as this method would reduce about 50% of the available flux capacity.

The excellent characteristics of the flux profile at the irradiation position in our reactor have provided us a better solution to achieve the desired axial uniformity without sacrificing the irradiation capacity. In this method two silicon crystals 20 cm long each, are irradiated together in an aluminium can, with their interface adjusted at the maximum flux position inside the irradiation tube. The irradiation tube is rotated about its axis at 2 rpm during irradiation. At precisely 50% of the total necessary dose, the crystals are pulled out of the core and their positions are interchanged as shown in fig. 3. The flux profile being symmetrical, the crystals now see almost mirror images of the respective profiles at previous positions. The irradiation then continues till the remaining dose is complete. Precise detection of the 50% fluence is the key to the success of this method. This is accomplished with the help of self-powered neutron detectors placed close to the

irradiation tube as well as the cobalt wire monitors which are irrradiated together with the silicon crystals.

The relation between the neutron dose ϕt [n/cm^2] received by the silicon crystals and the final resistivity ρ_f (Ω cm) attained after irradiation is given by [9]

$$\phi t = K\left[\frac{1}{\rho_f} - \frac{1}{\rho_i}\right], \qquad (2)$$

where ρ_i is the initial resistivity of the crystal, and

$$K = (N_0 \sigma_c \mu_e q_e)^{-1},$$

with

N_0 = number of atoms of ^{30}Si per cm^3 in the starting material,

σ_c = neutron capture cross section of ^{30}Si [cm^2],

μ_e = drift mobility of electrons in the crystal lattice [cm^2/V s],

q_e = electronic charge [C].

The constant K [Ω/cm] depends mainly on the neutron capture cross section of ^{30}Si and is characteristic of each reactor. Precise experimental determination of the value of K is essential for the calculation of the required neutron dose to achieve a given target resistivity.

The effective neutron dose received by the silicon crystals during the irradiation was measured by small cobalt wire monitors wrapped in thin aluminium sheets and fixed at the top and at the bottom of each crystal. The irradiation time is controlled by the integrated current signals from two self-powered neutron detectors (SPND) fixed very close to the irradiation tube.

After irradiation the aluminium can containing the crystals is removed from the irradiation tube and placed in the pool storage space away from the reactor core. The crystals remain in the storage for about 5 days to permit the decay of ^{31}Si ($T_{1/2} = 2.62$ h) and are then removed from the reactor, washed with distilled water, dried and monitored for residual radioactivity.

The doping process produces ^{32}P inside the silicon crystal through a secondary reaction when ^{31}P formed in the main transmutation reaction (1) continues to be irradiated with thermal neutrons. The ^{32}P decays with an emission of a β-particle and a half-life of 14.6 d. Although it exists in a very small quantity, the activity of this radionuclide must be measured and compared with the limits stated in the IAEA regulations for the safe transport of radioactive material before the crystals can be released for industrial processing. From the regulations, the level of ^{32}P radioativity with which the material could be classified as exempt is 7.4 Bq/g.

As the greater portion of the β-particles are self-absorbed within the crystal, to measure the radioactivity equivalent to 7.4 Bq/g is extremely difficult and it is much easier to relate the radioactivity to the count rate of β-particles. This involves careful calibration of the detector system.

Table 1
Calibration factors for the Geiger–Müller counting system

Calibration factor ($\times 10^{-2}$ cps g/Bq)	Type of absorber
2.079 ± 0.052	water (experimental)
2.029 ± 0.114	water (calculated)
2.094 ± 0.117	silicon (calculated)

The ^{32}P activity is monitored by means of a Geiger–Müller (G–M) detector which has been calibrated by using a ^{32}P standard aqueous solution with a geometry similar to that of silicon crystal. This was accomplished by placing the standard solution inside a PVC tube with a 4 in. diameter and 25 cm long. This tube was provided with windows made of 8 μm thick Macrofol KG film, where the G–M detector could be fitted. The radioactive ^{32}P solution was standardized in a 4π gas-flow proportional counter with an overall uncertainty of 2.5%. The calibration factor for the G–M detector is shown in table 1.

Since the experimental calibration of the G–M detector was made with aqueous solution instead of silicon, a simplified Monte Carlo code was developed to calculate the variation in the calibration factor in going from water to silicon. Details of this calculation will be published elsewhere. The results obtained for the calculated calibration factors are shown in table 1. The experimental and calculated values for the water medium agree with each other quite well. The difference betwen the water and silicon is also within the error of calculation, which is of the order of 5.6%. The results, therefore, indicate that the experimental factor can be used with confidence for the silicon crystals as well.

Each crystal is subject to check by the calibrated G–M detector. Different regions of the crystal surface are monitored separately, and the average value is compared with the IAEA limit.

6. Results of the test irradiations

A series of test irradiations was performed with float-zone silicon crystals having diameters of 3 and 4 in. and a length of 20 cm each. The silicon crystals were furnished by a Japanese manufacturer who shipped them usually in lots of four to six ingots. A total of 50 crystals were irradiated for the test experiments. The initial resistivity of the crystals varied from 1000 to 20 000 Ω cm, depending on the batch, and the target resistivities were 33, 54 or 110 Ω cm as specified in each case by the manufacturer. The irradiation of silicon crystals was carried out following the procedure outlined in the preceding section. After irradiation, the

crystals of each lot were checked for the residual radioactivity and finally shipped back to the manufacturer, where they underwent an appropriate thermal annealing process before measuring the final resistivity. The data on the final resistivity of each crystal in a given lot were furnished by the manufacturer along with the shipment of the next lot for the irradiation so that the irradiation parameters could be appropriately adjusted if necessary. The resistivity measurements were carried out on both flat faces of each ingot using a four point probe. The calculations of the axial and radial variation of the resistivity as well as the doping accuracy were made by using the following expressions:

1) Axial resistivity variation:

$$A = \bar{\rho}(U)/\bar{\rho}(L), \tag{3}$$

where $\bar{\rho}(U)$ and $\bar{\rho}(L)$ are the average resistivity values for the upper face (the crystal identification number is engraved on this face) and the lower face, respectively.

2) Radial resistivity variation:

$$R = \frac{\rho^{\max} - \rho^{\min}}{\rho^{\min}} \times 100, \tag{4}$$

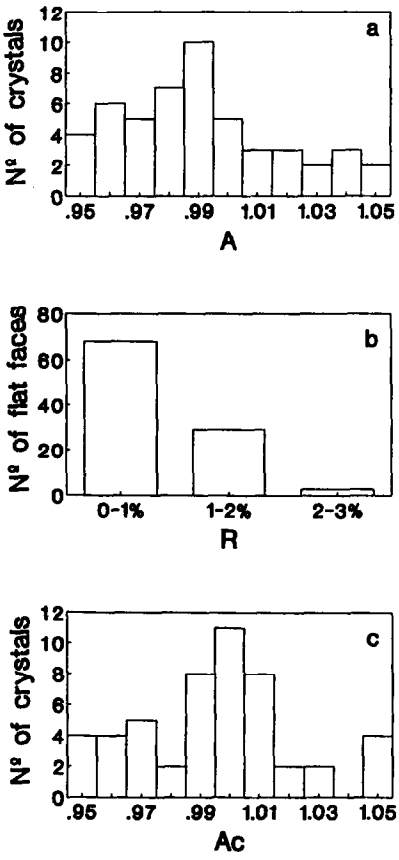

Fig. 4. Results of the test irradiations: Axial resistivity variation (a), radial resistivity variation (b) and doping accuracy (c). The quantities A, R and A_c are as defined in eqs. (3), (4) and (5) in the text.

where $\rho^{\max}$ ($\rho^{\min}$) are the maximum (minimum) resistivity values for each crystal face

3) Doping accuracy:

$$A_c = \bar{\rho}/\rho(T), \tag{5}$$

where $\bar{\rho}$ is the average final resistivity and $\rho(T)$ is the target resistivity.

The results obtained in the test irradiation are shown in fig. 4. These results clearly show the excellent doping quality achieved. About 70% of the crystals showed an axial variation of the resistivity equal or less than 3%. The maximum observed variation in all cases is 5% or less. Aproximately 70% of the crystals attained radial doping uniformity within 0 and 1%, while another 25% showed radial variation between 1 and 2%. In more than 70% of the cases the doping accuracy achieved was better than 3%. These results can be considered entirely satisfactory when compared with the commercially produced NTD silicon in other reactors.

Experimental data obtained in the test irradiations also served to determine a precise value of the calibration constant K apperaring in eq. (2). A least-squares fit of the data to eq. (2) gave, in the case of 4 in. crystals,

$$\phi t = (2.743 \pm 0.009) \times 10^{19}\left[\frac{1}{\rho_f} - \frac{1}{\rho_i}\right] \text{n/cm}^2. \tag{6}$$

7. Conclusion

A simple design irradiation rig has been constructed and installed in the IEA-R1 research reactor for the neutron transmutation doping of 3 and 4 in. diameter silicon crystals. A reasonably symmetric thermal neutron flux profile at the irradiation position permitted the irradiation of two silicon ingots, each 20 cm long, simultaneously. By interchanging the position of ingots at precisely half the required neutron dose, it has been possible to achieve an excellent axial doping uniformity. Test irradiation of 50 silicon crystals demonstrated excellent doping uniformities as well as doping precision. Considering the irradiation of only 4 in. crystals to a target resistivity of approximately 54 Ω cm, the irradiation capacity in our reactor is estimated to be at present of the order of 800 kg/y.

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