RADIAL UNIFORMITY OF NEUTRON IRRADIATION IN SILICON INGOTS FOR NEUTRON TRANSMUTATION DOPING AT HANARO

MYONG-SEOP KIM*, CHOONG-SUNG LEE, SOO-YOUL OH, SUNG-YUL HWANG and BYUNG-JIN JUN
Korea Atomic Energy Research Institute
150 Deokjin-Dong, Yuseong-gu, Daejeon, 305-353, Korea
*Corresponding author. E-mail : mskim@kaeri.re.kr

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The radial uniformity of neutron irradiation in silicon ingots for neutron transmutation doping (NTD) at HANARO is examined by both calculations and measurements. HANARO has two NTD holes named NTD1 and NTD2. We have been using the NTD2 hole for 5 in. NTD commercial service, and we intend to use two holes for 6 in. NTD. The objective of this study is to predict the radial uniformity of 6 in. NTD at the two holes. The radial neutron flux distributions inside single crystal and non-crystal silicon loaded at the NTD2 hole are calculated by the VENTURE code. For NTD1, the radial distributions of the reaction rate for a 6 in. NTD with a neutron screen are calculated by MCNP, and measured by gold wire activation. The results of the measurements are compared with those of the calculations. From the VENTURE calculation, it is confirmed that the neutron flux distribution in the single crystal silicon is much flatter than that in the non-crystal silicon. The non-uniformities of the measurements for radial neutron irradiation are slightly larger than those of the calculations. However, excluding local dips in the measurements, the overall trends of the distributions are similar. The radial resistivity gradient (RRG) for a 5 in. silicon ingot is estimated to be about 1.5%. For a 6 in. ingot, the RRG of a silicon ingot irradiated at HANARO is predicted to be about 2.1%. Also, from the experimental results, we expect that the RRG would not be larger than 4.4%.

KEYWORDS : Neutron Transmutation Doping (NTD), HANARO, RRG, Neutron Irradiation, Silicon

1. INTRODUCTION

Neutron transmutation doping (NTD) for producing n-type silicon semiconductor is based on the conversion of the Si-30 isotope into phosphorus atom by neutron absorption as follows,

$$Si^{30}(n,\gamma)Si^{31} \rightarrow P^{31} + \beta^{-} \ (T_{1/2} = 2.62 \text{ h}).$$

The resistivity of the semiconductor is inversely proportional to the P-31 atom concentration.

Using this doping method, silicon semiconductors with extremely uniform dopant distributions can be produced [1,2]; this is the main advantage of NTD compared with conventional chemical doping. Among the very wide applications of silicon semiconductors such as integrated circuits (IC), thyristors (SCR), transistors, etc., good uniformity of dopant concentration is generally required for high power applications in order to prevent hot spot formation and the eventual possibility of device breakdown [3,4] as well as for special sensors. NTD is one of the rare cases in research reactor utilization where direct industrial applications are possible [5].

HANARO, a 30 MW multipurpose research reactor, has two vertical holes in the heavy water reflector region for the NTD - NTD1 and NTD2 holes having diameters of 22 and 18 cm, respectively, as shown in figure 1. These two holes have excellent conditions for NTD from the viewpoints of high thermal neutron flux and large hole size for high throughput, and low fast neutron flux and gamma heating for low crystal defect [6,7]. Commercial NTD service for a 5 in. silicon ingot has been provided at the NTD2 hole. An additional 6 in. irradiation device at NTD2 and a 6 and 8 in. irradiation facility using the NTD1 hole are under development.

Uniform radial and axial irradiation in the ingot is the prime target in irradiation device design. The neutron screen method was chosen at HANARO to realize axially uniform irradiation. In this method, the appropriate neutron absorbing materials were used to obtain a uniform neutron flux distribution throughout the whole ingot. From a previous

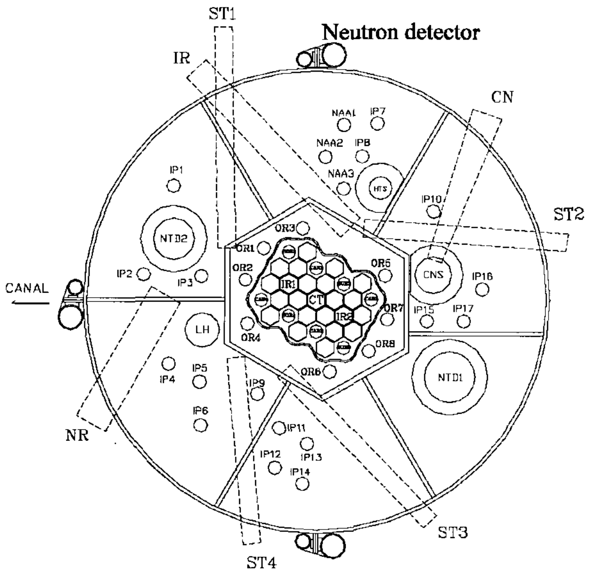

Fig. 1. Location of the NTD Holes at HANARO

study, the optimum neutron screen was established, and it provided an axial flux uniformity within ±2% for a 5 in. silicon ingot [8].

The radial uniformity of the resistivity in a semiconductor ingot can be represented by the RRG (radial resistivity gradient) as follows,

$$RRG = \frac{\rho_{max} - \rho_{min}}{\rho_{min}}, \quad (1)$$

where ρ_{min} and ρ_{max} are the minimum and maximum resistivity in a thin wafer, respectively. Wafer companies always require that the RRG be as low as possible, and their specifications are quite rigid. Because of the radial neutron flux gradient of the reactor, the silicon ingot must be rotated during irradiation in order to guarantee radial uniformity. Nevertheless, the RRG cannot be perfect because the neutron flux at the inner part of the silicon is slightly lower than that of the periphery due to neutron attenuation by the silicon, and the initial resistivity distribution is not uniform. The initial resistivity distribution cannot be reflected in the irradiation site. This non-uniformity becomes severer for ingots with larger diameter.

In this work, we investigated the radial uniformity of neutron irradiation for mainly 6 in. ingot by both calculations and measurements. First, the radial neutron flux distribution inside silicon material loaded at the NTD2 hole was calculated. This calculation was performed for a non-crystal silicon ingot as well as for single crystal in order to investigate the crystal effect. Then, the radial reaction rate distribution inside the 6 in. single crystal silicon ingot loaded at the NTD1 hole using the irradiation facility with a neutron screen was calculated and compared with the experimental results.

2. CALCULATION AND EXPERIMENTAL METHODS

VENTURE and MCNP were used to predict the radial distribution of neutron irradiation. We used the neutron cross-section of the silicon single crystal generated by improving the module LEAPR of the NJOY data processing system [9]. In the initial sensitivity calculations for the crystal and non-crystal silicon ingots, we used the VENTURE code to calculate the flux distributions inside the ingots loaded at the NTD2 hole where commercial service for a 5 in. silicon ingot has been ongoing. The optimum geometry of the neutron flux screen for the 5 in. NTD at the NTD2 hole was searched by the MCNP code because the geometry of the flux screen was too complicated to be dealt with properly by the node homogenization technique [8]. The optimum screen has provided excellent axial and radial uniformity for commercial NTD service. Therefore, a 6 in. irradiation device for the NTD1 hole was made on the basis of calculations using MCNP. We searched for an optimum neutron flux screen for the NTD1 hole giving the most uniform axial distribution of the $Si^{30}(n,\gamma)Si^{31}$ reaction rate.

Since our MCNP code does not yet have burnup calculation capability, all fuels have been assumed to be fresh. The effect of fuel burnup for axial uniformity was investigated by the HANARO fuel management system (HANAFMS) of WIMS and VENTURE, and it was concluded that the fuel burnup slightly affected the axial uniformity [8]. However, it is expected that the effect would be very small, because changes of the radial neutron flux distributions in the NTD holes according to the change of the core condition are very small as shown in figure 1. Since the general reliabilities of HANAFMS and MCNP have been confirmed many times in other works, recalculation by MCNP to estimate the radial uniformity at the NTD2 hole was not performed.

Figure 2 shows the VENTURE model for the irradiation of the silicon materials at the NTD2 hole. In order to simulate the neutronics of HANARO, a 3-dimensional H-Z model was adopted [10]. The pitch of a hexagonal cell is 1.1561 cm. The outside of the zircaloy tube is the D_2O region, and the silicon ingots are loaded at the center of the hole.

We assigned each ring a number, n=1, 2, 3, · · ·, and painted the cell with the same darkness. The thermal neutron flux at each cell was calculated, and the average neutron flux at each ring was obtained. For this calculation, the cross-section of single crystal silicon was added into the 69 group WIMS library [9]. The calculation was

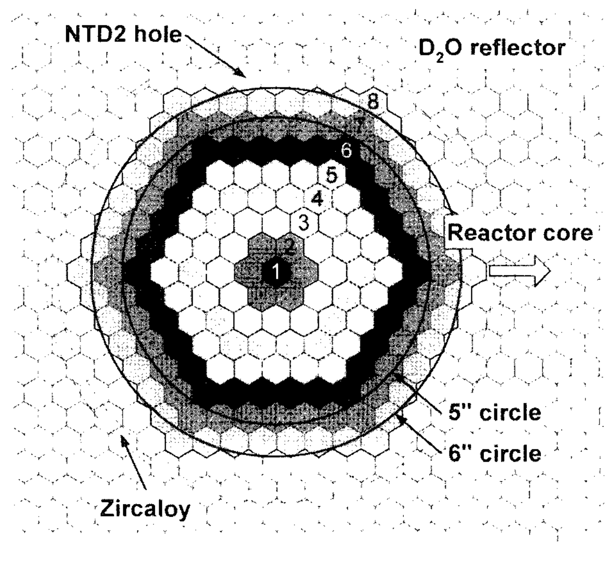

Fig. 2. Calculation Model for Irradiation of Silicon Loaded at NTD2 Hole

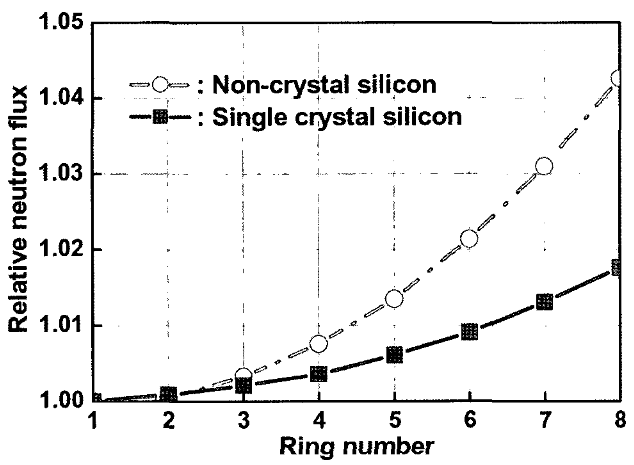

Fig. 3. Relative Neutron Fluxes at Each Hexagonal Ring Calculated by VENTURE

performed at the axial position of −10 ~ −15 cm from the longitudinal center of the reactor core, i.e., the height of a hexagonal cell was 5 cm.

In the MCNP calculation to obtain the radial $Si^{30}(n,\gamma)Si^{31}$ reaction rate distribution in the single crystal silicon ingot for the NTD1 hole, a model simulating a neutron screen with two pieces of 30 cm length ingots was used. The axial position of the ingot center in the reactor core was predetermined with consideration of the control rod position. We obtained data at three axial positions of a 60 cm silicon ingot – the top, bottom, and middle planes. The longitudinal length of each plane was 2 cm. The calculated reaction rates were averaged azimuthally in order to simulate the rotation of the ingot during irradiation.

The radial reaction rate distribution measurements were carried out using the irradiation facility made on the basis of MCNP calculations with 6 in. silicon ingots at the NTD1 hole. The measured data were compared with the calculation results. The gold wire activation method was used for the reaction rate distribution measurement [8]. Gold wire with a diameter of 0.1 mm was attached along the diameter on each plane of two pieces of 6 in. ingot having 30 cm length, using a thin aluminum tape. The ingots were irradiated at the NTD1 hole for 1 hour at a reactor power of 30 kW. The ingots were rotated at a speed of 5 rpm. The 411.8 keV gamma-ray from the irradiated gold wire was measured using a high purity germanium detector by the gamma-scanning method. The saturated activity distributions were determined by considering the decay time. The lead slit size of the gamma-scanning system was 4 mm.

3. RESULTS AND DISCUSSIONS

Figure 3 shows the relative neutron fluxes computed by VENTURE at each hexagonal ring depicted in figure 2. The neutron flux at each ring was obtained by averaging the cell fluxes in each ring in order to consider the rotation of the silicon ingot. The differences in the neutron flux between ring 1 (center) and ring 8 are 4.3% for non-crystal silicon and 1.8% for single crystal silicon, respectively.

Because the geometry is hexagonal, the distances from the NTD2 hole center to the cells with the same ring number are not the same. The maximum difference of distances is about 11%. Hence, the ring number in figure 3 does not exactly correspond to the radius of the silicon ingot. Table 1 shows the neutron fluxes for a few rings calculated using VENTURE.

From figure 2, we find that the average distance from the ingot center to the 7^{th} ring is about 5 in. Therefore, the neutron flux at the 7^{th} ring would be very similar to the peripheral flux of the 5 in. silicon ingot, and that at the 8^{th} ring would be slightly smaller than the peripheral flux of the 6 in. ingot. From above results, the radial uniformities of resistivity are expected to be less than 1.5 and 2% for 5 and 6 in. silicon single crystal ingots, respectively, without initial resistivity distribution. If the ingots are not single crystal, theses values become larger by 2~3 times.

The total cross-section of single crystal silicon is smaller than that of non-crystal silicon for neutron energy below several hundred meV [11,12]. It is clear that the reduced neutron scattering cross-section of the single crystal silicon makes the ingot more transparent to neutrons and gives a much more uniform neutron flux distribution than that in

Table 1. Neutron Fluxes Calculated Using VENTURE for a Few Rings of the Silicon Ingot

	Average radial distance [cm]	Non-crystal silicon		Single crystal silicon	
		Neutron flux, absolute [n/cm²s]	Neutron flux, relative	Neutron flux, absolute [n/cm²s]	Neutron flux, relative
Center	0	5.258×10^{13}	1.000	5.135×10^{13}	1.000(0.977*)
Average at 7th ring	6.31	5.421×10^{13}	1.031	5.202×10^{13}	1.013
Average at 8th ring	7.36	5.482×10^{13}	1.043	5.226×10^{13}	1.018(0.953*)

* Ratio of fluxes for single crystal silicon to non-crystal silicon

the case of the non-crystal silicon. It also increases neutron leakage, and therefore the neutron flux level in and around the NTD2 hole is decreased. The neutron flux at the central hexagon of single crystal silicon is about 2.3% lower than that of non-crystal silicon.

In this calculation, the neutron cross-sections at a temperature of 300 K were used. However, in the actual irradiation of the ingot at a reactor power of 20~30 MW, the ingot temperature becomes higher due to radiation heating. Although the absorption cross-section is almost temperature-independent, the scattering cross-section of a single crystal is larger at higher temperature. Hence, the radial non-uniformity of the neutron flux distribution in the actual doping process is expected to be slightly larger than that of the calculation.

Figure 4 shows the relative $Si^{30}(n,\gamma)Si^{31}$ reaction rates calculated by MCNP as a function of the radial distance from the center of a 6 in. silicon crystal ingot loaded at the NTD1 hole. The relative neutron flux distribution inside silicon loaded at the NTD2 hole obtained by VENTURE is also represented in the same figure. The equivalent radial distance of the VENTURE result was deduced by finding the circle of the area equal to that of each hexagonal ring.

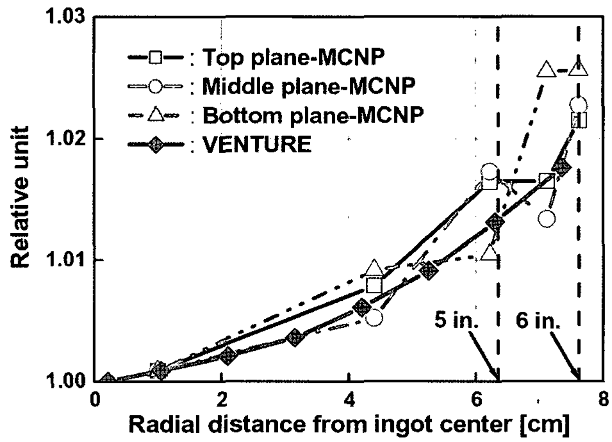

Fig. 4. Relative Radial $Si^{30}(n,\gamma)Si^{31}$ Reaction Rates Calculated by MCNP for 6 in. Silicon Crystal Ingot Loaded at the NTD1 Hole and Relative Neutron Flux Obtained by VENTURE for the NTD2 Hole

The MCNP model simulated the actual irradiation geometry almost exactly. The calculated values at each plane shown in the figure were normalized by the surface averaged values at a 1 cm radius. In order to compare these results with those obtained using VENTURE, the appropriate offset was applied to the MCNP results. The difference between the axial reaction rates at the center of each plane was confirmed to be less than 1%.

It is confirmed that the calculated reaction rate is lowest at the center and highest at the periphery of the ingot. The differences between them are less than 2.5%. The fractional standard deviation (fsd) of the calculated reaction rate is 0.7% at the periphery, and 2% at the radius of 1 cm.

The NTD irradiation facility of HANARO has neutron reflecting materials above and below the position of the silicon ingot in order to increase the neutron flux at both ends of the ingot. Because of these materials, distortions in the radial neutron flux distributions at the top and bottom planes of the ingot might occur. However, since the radial reaction rate distributions at three planes are similar, the effect of the neutron reflecting materials on the radial reaction rate distribution is considered negligible. From figure 4, it is confirmed that the 1% criterion for the radial resistivity variation noted in a previous work [13] is difficult to achieve for ingots with a radius larger than 4 in. even if the initial resistivity distribution is ignored.

Although two calculations are very different in the calculation tool (VENTURE and MCNP), geometrical modeling (hexagonal node homogenization and almost actual geometry), irradiation position (NTD2 and NTD1) and parameters in comparison (thermal neutron flux and Si-30 neutron capture reaction rate), the difference between the results of two calculations is very small compared to the fsd of MCNP calculation.

From the previous MCNP calculation for the axial direction, it was confirmed that the difference between the normalized axial distribution of the neutron flux and that of the Si-30 reaction rate was a few %. This difference is attributed to the difference in the neutron spectrum depending on the axial position [8]. However, this difference is not expected in the radial distribution even though the neutron spectra at the two holes are different, because the Cd ratio of gold in the irradiation area of the ingot is about 20 and its spatial discrepancy in a plane is very small [8].

Figure 5 shows the relative radial reaction rates measured by gold wire activation in the irradiation experiment for a 6 in. ingot at NTD1 and the neutron flux distribution calculated by VENTURE. Two results were normalized by the values at the Si ingot center. For the purpose of comparison, the measured results were moved parallel so that the results of the VENTURE calculation would fit the measurements. The difference between the measured values at the center of each plane, i.e. the axial non-uniformity, was less than 1%, which agreed very well with the prediction by the MCNP calculation.

The difference between the measurements in the radial direction was slightly larger than that in the calculation, and the maximum value was 4.4%. The uncertainty in the determination of the gamma-ray peak area was smaller than 1.0%.

From the figure, excluding local dips in the measurements, the overall trends of the distributions are similar. The effect of the difference in the energy dependency of the cross-section between Au-197 and Si-30 is negligible. In addition to the statistical uncertainty in the peak area, we should take into account uncertainties arising from the non-uniformity of the Au wire diameter and possible bending of the wire. However, these uncertainties are not easily determined. Also, attaching the Au wire using thin aluminum tape on the surface of the ingot results in a thin water layer, having a thickness of at least 0.2 mm, between the ingot surfaces. Since water is a relatively strong neutron attenuator, it may have some effect on the measured radial uniformity. When the RRG distributions along the axial direction of the ingots were measured in a test irradiation for a 5 in. silicon ingot at the NTD2 hole, the RRG tended to become larger at positions closer to the end of the ingot, which could be explained by the effect of the thin water layer. However, since only a single case was analyzed in this regard, further study is required for verification of this phenomenon.

In 5 in. NTD commercial service at the NTD2 hole, RRG records measured by customers show a distribution between 0.4 to 4.2%, with an average of 2.05% and a standard deviation of 0.82%. The distribution is originated from the initial resistivity distribution, uncertainty in the resistivity measurement, and deviation from ideal irradiation conditions.

From the above results, we estimated the RRG for 5 in. ingot to be about 1.5%. The increase of RRG due to the increase of ingot diameter from 5 to 6 in. is estimated only from the results of the VENTURE calculation, i.e., approximately 0.6%. Therefore, for a 6 in. ingot, the RRG of a silicon ingot irradiated at HANARO is predicted to be about 2.1%. Also, from the experimental results, we expect that the RRG for 6 in. silicon ingot would not be larger than 4.4%, which might be acceptable.

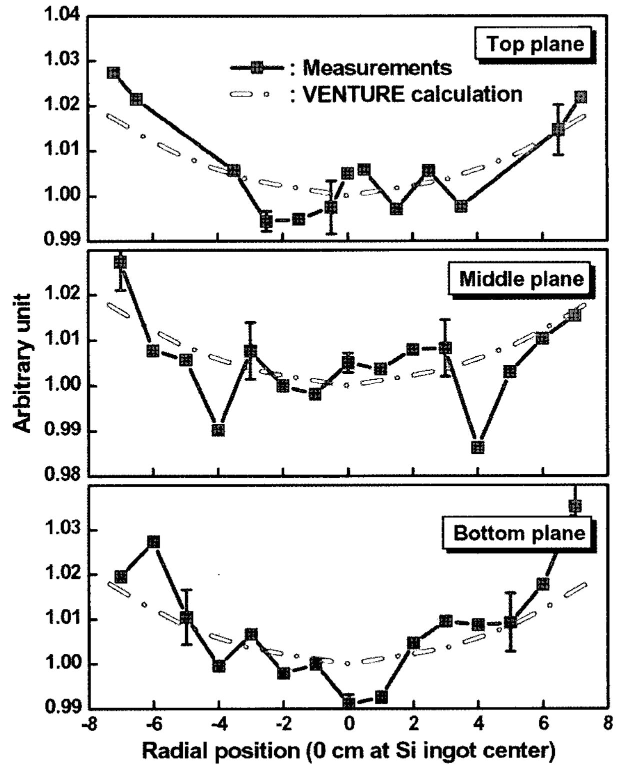

Fig. 5. Comparisons of the Measured Radial Reaction Rate Distributions at the NTD1 Hole and the Neutron Flux Distribution Calculated by VENTURE for the NTD2 Hole

4. CONCLUSION

We have confirmed the radial uniformity of neutron irradiation for NTD of silicon by calculations and measurements. The RRG of a 6 in. silicon ingot irradiated at HANARO is predicted to be about 2.1%. From the experimental results, we expect that the RRG would not be larger than 4.4%, which might be acceptable.

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