NEW EVALUATION METHODS FOR RADIAL UNIFORMITY IN NEUTRON TRANSMUTATION DOPING

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Recently, the neutron irradiation for large diameter silicon (Si)-ingots of more than 8" diameter is requested to satisfy the demand for the neutron transmutation doping silicon (NTD-Si). By increasing the Si-ingot diameter, the radial non-uniformity becomes larger due to the neutron attenuation effect, which results in a limit of the feasible diameter of the Si-ingot. The current evaluation method has a certain limit to precisely evaluate the radial uniformity of Si-ingot because the current evaluation method does not consider the effect of the Si-ingot diameter on the radial uniformity. The objective of this study is to propose a new evaluation method of radial uniformity by improving the conventional evaluation approach. To precisely predict the radial uniformity of a Si-ingot with large diameter, numerical verification is conducted through comparison with the measured data and introducing the new evaluation method. A new concept of a gradient is introduced as an alternative approach of radial uniformity evaluation instead of the radial resistivity gradient (RRG) interpretation. Using the new concept of gradient, the normalized reaction rate gradient (NRG) and the surface normalized reaction rate gradient (SNRG) are described. By introducing NRG, the radial uniformity can be evaluated with one certain standard regardless of the ingot diameter and irradiation condition. Furthermore, by introducing SNRG, the uniformity on the Si-ingot surface, which is ignored by RRG and NRG, can be evaluated successfully. Finally, the radial uniformity flattening methods are installed by the stainless steel thermal neutron filter and additional Si-pipe to reduce SNRG.

KEYWORDS : Neutron Transmutation Doping (NTD), Radial Uniformity, Radial Resistivity Gradient (RRG), Normalized Reaction Rate Gradient (NRG), Partial Normalized Reaction Rate Gradient (PNRG), Surface Normalized Reaction Rate Gradient (SNRG)

1. INTRODUCTION

The neutron transmutation doping silicon (NTD-Si) for n-type semiconductors is produced by the conversion of ^{30}Si isotope, whose abundance is 3.12% in natural silicon, into a phosphorus atom by the neutron absorption reaction as follows:

$$^{30}\mathrm{Si}\,(\,n,\gamma\,)\,^{31}\mathrm{Si} \;\rightarrow\; ^{31}\mathrm{P} + \beta^- \quad (T_{1/2} = 1.26\ \mathrm{h}).$$

The resistivity of a semiconductor is inversely proportional to the ^{31}P atom concentration. Using this method, silicon semiconductors with uniform resistivity distribution can be produced [1]. The uniform resistivity distribution is the main advantage of NTD compared with conventional chemical doping methods. This uniform resistivity distribution is generally required to produce high power semiconductors and special sensors to decrease the possibility of device breakdown [2, 3]. Then, a prime target of NTD is to achieve uniform neutron irradiation throughout an entire Si-ingot since the resistivity distribution mainly depends on the neutron irradiation. While NTD was firstly launched at research reactors using a 2" diameter silicon (Si)-ingot in the mid-1970s, the Si-ingot diameter had been gradually increased year after year [4]. Nowadays, the worldwide demand for high power semiconductors is rapidly increasing in view of the sufficient supply for new power generation systems, such as hybrid cars and hydrogen fuel cell engines. In order to satisfy the supply of high power semiconductors, larger diameter Si-ingot irradiation was requisite [5].

The resistivity distribution caused by the neutron attenuation effect was found inside the Si-ingot even though the silicon itself was quite transparent to the neutrons. Since the Si-ingot for NTD was a cylindrical shape, its irradiation uniformity was generally expressed by the axial and radial variation of resistivity after the irradiation. The axial irradiation uniformity, which was caused by the axial neutron flux distribution, was achieved by conventional flattening methods, including the reverse, round-trip and fixed methods used in many research reactors [6]. The radial irradiation uniformity became almost flattened in the Si-ingot with small diameter since the neutron mean free path is sufficiently long [7]. However, in the Si-ingot with large diameter, the radial irradiation uniformity became worse because of the neutron attenuation effect. Then additional efforts were concentrated on the radial uniformity flattening for the Si-ingot with large diameter.

The current evaluation method had a certain limit to precisely examine the radial uniformity of the Si-ingot because the current evaluation method did not consider the effect of Si-ingot diameter on the radial uniformity. With increasing Si-ingot diameter, the limitations, which correspond to the allowable difference among resistivity distributions, were actually applied. Even though various limits were applied to the Si-ingot with a different diameter, the NTD semiconductor was expected to maintain an excellent advantage of NTD, i.e., very high uniformity. Additionally, by increasing the Si-ingot diameter, the curvature of the resistivity distribution becomes distorted with a steeper slope since the resistivity distribution inside the Si-ingot has an exponential-like shape due to the neutron attenuation effect. This tendency would introduce the non-uniform performance of each semiconductor device cutting from a Si-wafer since the resistivity would be changed and would depend on the position inside the Si-wafer.

From the viewpoint of radial uniformity, in this study, attention was paid to the effects of the change in Si-ingot diameter and Si-wafer position dependence. The objective of this study is to propose a new evaluation method of radial uniformity in NTD in order to improve conventional approaches, such as RRG.

2. NUMERICAL VERIFICATION

HANARO (a 30 MW multi-purpose research reactor in the Republic of Korea) has two vertical irradiation holes with 22- and 18-cm diameters for the NTD service in the heavy water reflector region, as shown in Fig. 1. The NTD service for 6" and 8" Si-ingot irradiation can be conducted using the two irradiation holes. For the uniform neutron irradiation of the entire Si-ingot, the neutron screen method was applied to realize an axial by uniform irradiation and the Si-ingot was rotated to achieve a radial uniform irradiation [8]. In these methods, the appropriate neutron absorption materials were used for the neutron screen to obtain a uniform reaction rate distribution throughout the entire Si-ingot. After the irradiation, the radial irradiation uniformity was checked by measuring the radial reaction rate distribution using the gold wire activation method [9]. Gold wire 30 cm in length was attached along the radial direction on each plane of two pieces of 6" and 8" Si-ingot. The 6" and 8" Si ingots were irradiated for one hour with 30 kW at HANARO. The 411.8 keV γ-ray from the irradiated gold wire was measured using a high purity germanium detector.

Recently, a lager diameter Si-ingot irradiation above 8" was requested to satisfy the demand for NTD-Si. However, increasing the Si-ingot diameter, the radial non-uniformity became larger due to a neutron attenuation effect, and it resulted in the limit of the feasible diameter of the Si-ingot. The maximum diameter of the Si-ingot available in a commercial NTD service was 8", and these experimental data of radial uniformity of 8" Si-ingot were not opened because this information is kept confidential in the industry. For these reasons, a verification of numerical analyses was required to precisely predict the radial uniformity for Si-ingots larger than 8" diameter.

In order to examine the feasibility of numerical analyses,

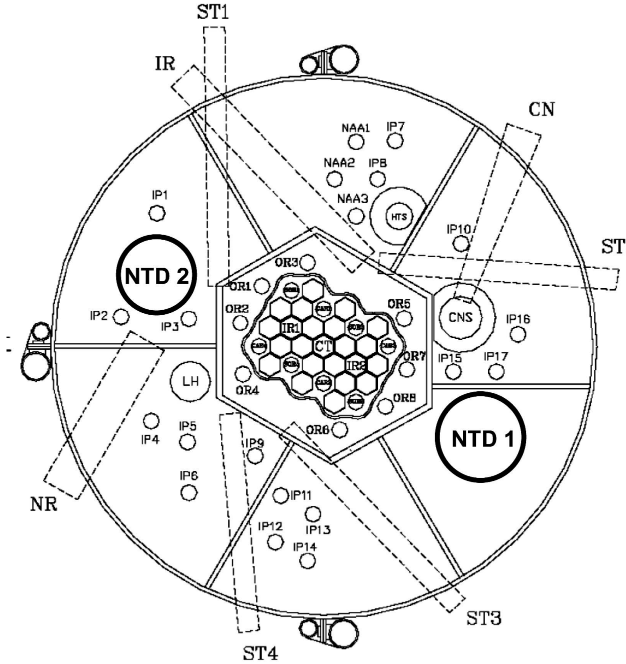

Fig. 1. Location of NTD 1 and 2 holes in the Reflector of HANARO

the radial uniformity of a 6" Si-ingot was numerically evaluated by MCNPX-2.6 [10] code with the ENDF/B-VI.2 [11] nuclear data library, and these numerical results were compared with the measured ones. For profiling the radial reaction rate distribution, the 6" Si-ingot was radially divided by 0.5" concentric rings, and the reaction rate in the each concentric ring was averaged due to consideration of the rotation effect of Si-ingot. A series of MCNPX calculations were executed by a total of 5×10^7 histories, and the fractional standard deviation (fsd) of ^{30}Si(n, γ)^{31}Si reaction rates were less than 0.5%.

In the MCNPX calculations, the neutron cross-section of single crystal silicon generated by the improved module, LEAPR, of NJOY was employed [12, 13]. Figure 2 shows the difference of the neutron total cross sections between single crystal and non-crystal silicon. The total cross-section of single crystal silicon was smaller than that of non-crystal silicon for neutron energy less than 0.5 eV because of the fewer neutron scattering reactions [14], and the neutron attenuation by the small neutron scattering cross-section made the Si-ingot more transparent to neutrons. Because of this, the radial uniformity by adopting the cross-section of single crystal silicon became flatter than that by the non-crystal silicon, which indicates that the proper usage of the silicon cross-section is important to evaluate the radial uniformity of especially large diameter Si-ingots.

Figure 3 shows the comparison of the relative radial reaction rate distributions between the calculated and measured results of gold wire reaction rates in a 6" Si-ingot. The overall trends of measured and calculated radial reaction rate distributions showed good agreement except for a few points, and the maximum difference between the measured and calculated results was within 2%. Through this comparison, the numerical evaluation by MCNPX calculations using the single crystal silicon cross-section and ENDF/B-VI.2 was confirmed to be feasible.

3. NEW EVALUATION METHODS FOR RADIAL UNIFORMITY

3.1 Current evaluation method

The radial resistivity uniformity of an NTD silicon wafer was conventionally evaluated by the radial resistivity gradient (RRG) as follows:

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

where ρ_{max} and ρ_{min} indicate the maximum and minimum resistivities in a layer of a silicon wafer, respectively. The resistivity of silicon semiconductor was decided by the amount of ^{30}Si(n, γ)^{31}Si reaction, and the radial irradiation uniformity was conventionally evaluated by substituting the maximum and minimum resistivities into ^{30}Si(n, γ)^{31}Si

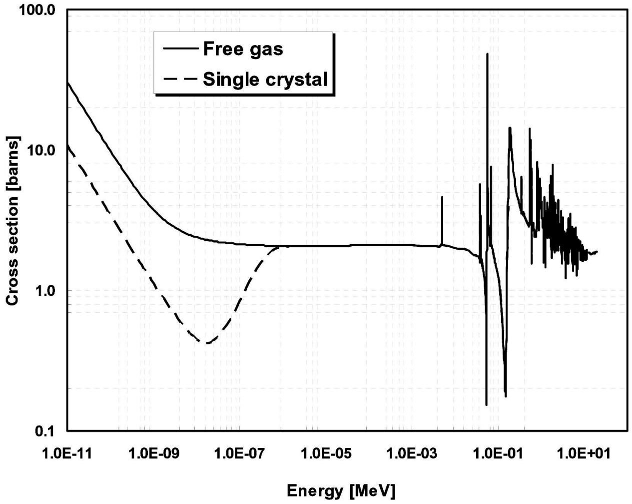

Fig. 2. Comparison of Neutron Total Cross-section Between Single and Free Gas Silicon

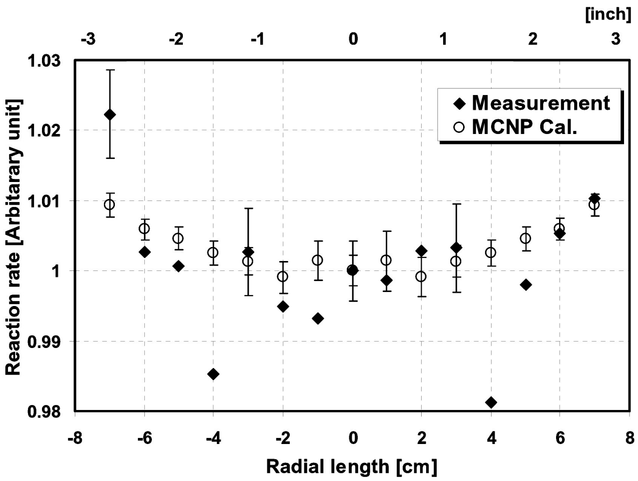

Fig. 3. Comparison of the Radial Reaction Rate Distribution Between the Measurement and Calculation in 6" Si-ingot

Table 1. Radial Uniformity Limit and Calculated RRG Results Varying Si-ingot Diameter

Ingot diameter (inch)	RRG limit (%)	RRG (%)
6	4	2.2
8	5	3.9
10	-	5.8
12	-	7.7
14	-	9.8

reaction rates. Then, RRG in Eq. (1) can be rewritten as follows:

$$RRG = \frac{RR_{max} - RR_{min}}{RR_{min}}, \qquad (2)$$

where RR_{max} and RR_{min} are the maximum and minimum $^{30}Si(n, \gamma)^{31}Si$ reaction rates, respectively. In Eqs. (1) and (2), RRG is defined by the difference between the maximum and minimum values normalized by the minimum value. Through the normalization, the radial uniformity could be evaluated regardless of the irradiation conditions, including neutron flux level and irradiation time. However, with increasing Si-ingot diameter, the difference between the maximum and minimum reaction rates becomes larger because of the neutron attenuation effect. Therefore, the value of RRG becomes larger with increasing Si-ingot diameter.

The current evaluation method (RRG) evaluated the radial uniformity with a concept of difference between the maximum and minimum reaction rates, and a different limit of radial uniformity was applied to each Si ingot's diameter, where the limit was provided by semiconductor companies. Table 1 shows the current radial uniformity limit of various Si-ingot diameters. The RRG value shows an increasing tendency with increasing Si-ingot diameter, and it is clear that the radial uniformity cannot be evaluated by one limit. From these results, the use of these different limits is considered to be difficult to provide the information about the effect of ingot diameter increase on the radial uniformity. This implication comes from the fact that the effect of Si-ingot diameter is not considered in RRG.

3.2 New concept

A new approach to deal with the radial uniformity, instead of the current RRG interpretation, is described in this sub-section. As shown in Fig. 4, the maximum reaction rate was found at the ingot surface, while the minimum reaction rate was found at the center of the Si-ingot since the radial reaction rate distribution is clearly related to the neutron attenuation effect. Here, a concept of gradient between the surface and center of the ingot, called the normalized reaction rate gradient (NRG) was newly introduced, as follows:

$$\begin{aligned} NRG &= \left[\quad Gradient \quad \right] \times \begin{bmatrix} Normalization \\ factor \end{bmatrix} \\ &= \left[\frac{RR_{surface} - RR_{center}}{r} \right] \times \left[\frac{1}{RR_{center}} \right], \end{aligned} \qquad (3)$$

where $RR_{surface}$ and RR_{center} indicate the $^{30}Si(n, \gamma)^{31}Si$ reaction rates at the surface and center of the Si-ingot, respectively, and r is the ingot radius. In Eq. (3), the difference between the reaction rates at the surface and center can be interpreted as a gradient in consideration of the ingot radius, and the normalization is applied in the same way as RRG by using the minimum reaction rate. Using this normalized gradient, Eq. (3) can be illustrated as in Fig. 4. The thick

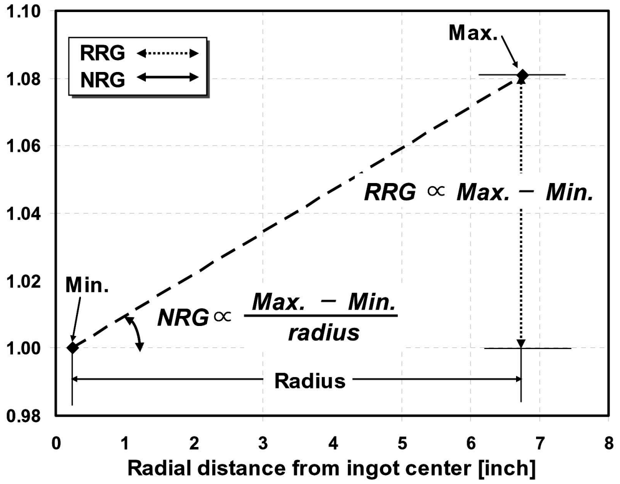

Fig. 4. Concept of the RRG and the NRG for Radial Uniformity Evaluation

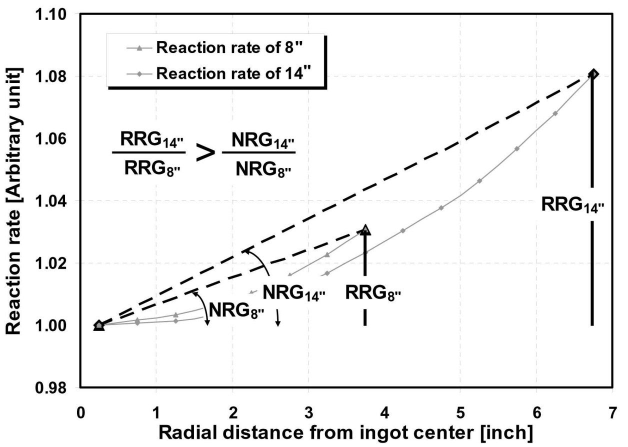

Fig. 5. Different Evaluation of the Effect of Si-ingot Diameter Increasing in RRG and NRG

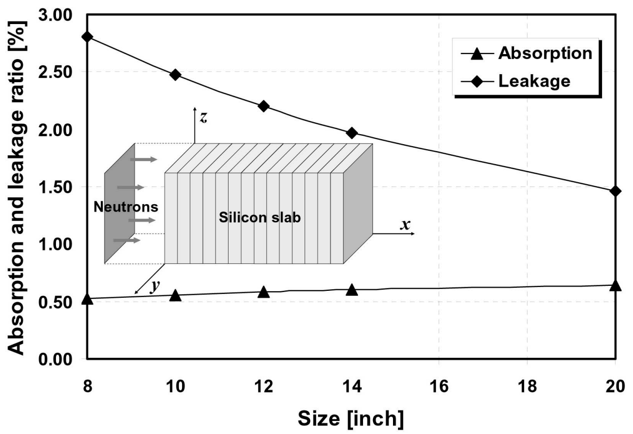

Fig. 6. Comparison Between Neutron Leakage and Absorption in Different Size of Slab Geometries

Table 2. Comparison of the Radial Uniformity Calculated by RRG and NRG

Ingot diameter (inch)	RRG (%)	NRG (%/inch)
6	2.2	0.7
8	3.9	1.0
10	5.8	1.2
12	7.7	1.3
14	9.8	1.4

solid line parallel to the y-axis corresponds to the RRG value, and an angle between the dotted line and the base line corresponds to the NRG value. This gradient can represent the resistivity distribution, and an equivalent gradient of different Si-ingot diameters means that each Si-ingot has an equivalent resistivity distribution.

In order to show an effect of NRG introduction by Eq. (3), the radial uniformity of various diameters of Si ingots was compared in the values of RRG and NRG. MCNPX was employed in the numerical simulation of irradiation for Si ingots with 6" to 14" diameters in the HANARO reflector region. In these calculations, a neutron screen employed to achieve the axial uniformity was removed in order to make a simple comparison between RRG and NRG, whereas the neutron screen was included in actual irradiation service. As shown in Table 2 and Fig. 5, the increase in RRG value is approximately three times larger than that of NRG when the Si-ingot diameter was increased from 6" to 14". The fact shows that the change in NRG value with increasing ingot diameter was very different from that in RRG; the change of NRG value was considerably smaller than that of RRG. From these results, it is clear that the NRG concept is usable in the evaluation of radial uniformity for the consideration of the effect caused by the Si-ingot diameter change.

3.3 Analysis of the Si-wafer radial non-uniformity in large ingots

In Sec. 3-2, the radial uniformity was obtained using only the maximum and minimum reaction rates even though the reaction rate distribution had a curvature as shown in Fig. 5. This curvature of the radial reaction rate distribution could be easily analyzed in small diameter Si-ingots, such as 2"; however, it is difficult to apply NRG directly to a large diameter Si-ingot since the NRG value cannot express the actual reaction rate distribution inside the Si-ingot, as indicated in Fig. 5.

In order to figure out the feature of curvature of the radial reaction rate distribution dependent on the ingot diameter, the neutron attenuation effect inside silicon was investigated using a simplified calculation model of slab geometry for the 0.1 eV mono-energy and mono-directional neutron beam. Figure 6 shows the comparison between the neutron leakage and absorption ratio when the size of silicon slabs was extended to the y direction from 8" to 20" with an assumption of an infinite slab to the *x* and *z* direction. While increasing the silicon slab size, the neutron leakage from the silicon slab surface is decreased, whereas the neutron absorption is slightly increased and the ratio of neutron leakage is always larger than that of neutron absorption. From these results, the neutron leakage by scattering is considered to have a larger effect than the neutron absorption for the neutron attenuation in silicon. The combination of neutron leakage at the surface and the neutron absorption inside the Si-ingot is considered to the cause of the formation of the curvature of the reaction rate distribution, and finally the curvature becomes larger with increasing Si-ingot diameter.

3.4 Application to Si-wafer radial non-uniformity

To express the distribution inside the Si-wafer, the new concept described in Sec. 3-2 was applied to the evaluation of partial gradient in the radial reaction rate distribution view of utilization of small pieces. Figure 7 illustrates the concept of small piece evaluation by using the differential of reaction rate distribution. The concept of partial slope was newly introduced to evaluate the radial uniformity in view of producing small size semiconductor device, which is called the partial normalized reaction rate gradient (PNRG) and is defined as follows:

$$PNRG = \begin{bmatrix} Gradient \\ inside\ a\ piece \end{bmatrix} \times \begin{bmatrix} Normalization \\ factor \end{bmatrix}$$

$$= \left[\frac{RR_{r_i} - RR_{r_{i-1}}}{\Delta r}\right] \times \left[\frac{1}{RR_{r_{i-1}}}\right] \qquad (\,i=2,3,4 \cdots\cdots n), \tag{4}$$

where RR_{r_i} and $RR_{r_{i-1}}$ indicate the maximum and minimum $^{30}Si(n, \gamma)^{31}Si$ reaction rates inside the *i*-th silicon piece and Δr is the length of the *i*-th silicon piece. In Eq. (4), the difference between the maximum and minimum reaction rates inside of the silicon piece was considered to be the gradient inside of the silicon piece by dividing the size of the silicon piece, Δr. Then, the gradient was normalized by the minimum reaction rate inside the piece.

Table 3. Evaluation of the Uniformity Distribution Inside a Si-Wafer using PNRG

Ingot diameter (inch)	$PNRG_2$ (%/inch) - center -	$PNRG_n$ (%/inch) - surface -
8	0.79	1.83
10	0.77	2.08
12	0.72	2.43
14	0.64	2.94

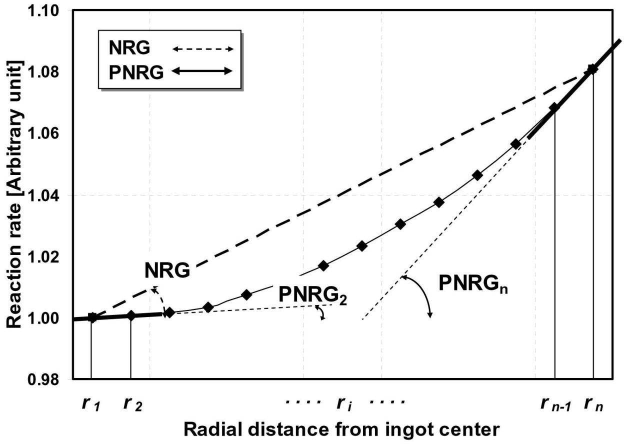

Fig. 7. Concept of NRG and PNRG for Radial Uniformity Evaluation

In order to examine the feasibility, the position dependence of the PNRG inside the Si-wafer was investigated. As shown in Table 3, the maximum gradient, $PNRG_n$, of each Si-ingot was approximately two times larger than the Normalized Reaction rate Gradient (NRG). Moreover, when the Si-ingot diameter was increased from 8" to 14", the minimum gradient ($PNRG_2$) at the ingot center region decreases by 20% and the maximum gradient ($PNRG_n$) at the ingot surface region increases by 60%. This fact indicates that the quality of each semiconductor piece produced from the irradiated Si-wafer considerably varied with increases in the Si-ingot diameter. Therefore, it is requisite to consider PNRG, especially to realize the radial uniformity of a semiconductor device through the neutron irradiation of a Si-ingot with large diameter.

To express the regional effect more precisely, a modified evaluation method was proposed to analyze the possible radial uniformity of semiconductor device at the outermost region where the non-uniformity generally reaches its maximum. Then, the concept of surface normalized reaction rate gradient (SNRG) was proposed by the value of the maximum PNRG as follows:

$$SNRG = Max.\left[PNRG\right] = \left[\frac{RR_{r_n} - RR_{r_{n-1}}}{\Delta r}\right] \times \left[\frac{1}{RR_{r_{n-1}}}\right], \tag{5}$$

where RR_{r_n} and $RR_{r_{n-1}}$ are the maximum and minimum $^{30}Si(n, \gamma)^{31}Si$ reaction rates inside the outermost silicon piece of Si-wafer, respectively, and Δr is the size of the silicon piece. In Eq. (5), SNRG is the maximum value of PNRG, and SNRG is specialized to evaluate the uniformity of the outermost silicon piece having the least uniformity. By introducing SNRG, the uniformity distribution inside the Si-wafer could be evaluated easily. If the worst uniformity of the silicon piece at the ingot surface could satisfy the limit of radial uniformity, other silicon pieces could naturally satisfy the limit. And, when the value of SNRG exceeds the limit, this non-uniformity could be adjusted by reducing the SNRG value below the limit.

In order to achieve the radial uniformity by reducing the value of SNRG, two flattening methods were considered as a preliminary step. The first method uses a thermal neutron filter for neutron spectrum hardening. The reduced neutron scattering by the harder neutron spectrum should make a Si-ingot more transparent, and the radial reaction rate distribution could be flat. Here, stainless steel, which was used as a neutron screen to achieve axial uniformity in HANARO, was chosen as a thermal neutron filter. Then, the axial and radial uniformity could be achieved using a properly designed neutron screen. The second method is to add a silicon pipe outside the Si-ingot, and the Si-ingot inside the silicon pipe can be achieved flat radial uniformity by removing the additional silicon pipe after irradiation. The advantage of this additional Si-pipe was to maximize the neutron flux inside the Si-ingot by minimizing the effect of thermal neutron filtering, and the minimized loss of neutron flux could reduce the total irradiation time for achieving a target resistivity. Table 4 shows the effect of these two flattening methods for reducing SNRG. From the results, the SNRG values are reduced by more than 45% and 30% by introducing the stainless steel filter and additional silicon pipe, respectively, and it was confirmed that the radial uniformity was successfully achieved by reducing the value of SNRG.

Table 4. Effect of SNRG using a Stainless Steel Thermal Neutron Filter and an Additional Silicon Pipe

Ingot diameter (inch)	SNRG (%/inch)		
	Bare	Stainless steel screen	Silicon pipe
8	1.83	0.82	1.08
10	2.08	1.09	1.47
12	2.43	1.38	1.76
14	2.94	1.63	2.14

4. CONCLUSION

A new evaluation concept in consideration of the gradient of reaction rate distribution was proposed for the analyses of NTD since a drawback of the current evaluation method (RRG) was found by increasing the Si-ingot diameter. Using the new concept of gradient (NRG), the radial uniformity was evaluated with a certain standard, regardless of ingot diameter and irradiation condition. The PNRG evaluation based on partial NRG was applied to a small size piece inside a Si-wafer in order to evaluate the local position effect caused by the radial reaction rate distribution. Furthermore, PNRG was extended to SNRG, which aims to deal with the worst value of radial distribution generally observed at the ingot surface. Introducing SNRG, the radial uniformity, even in a small area at Si-ingot surface, can be treated exactly. Finally, the installation of a stainless steel thermal neutron filter and additional silicon pipe methods were very effective in radial uniformity flattening by reducing SNRG. In the future, two flattening methods based on the NRG and SNRG concepts will be studied more precisely to achieve the radial uniformity both in large and small pieces of Si-wafer and Si-device, respectively.

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