Simulation Analysis of p-type Gallium Oxide Preparation by Proton Irradiation Transmutation Doping

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Abstract

Ultra-wide bandgap semiconductor gallium oxide is currently a material of intense research interest in the semiconductor field; however, p-type doping of bulk crystals has not yet been achieved through conventional doping processes, which hinders its practical applications. Proton irradiation transmutation doping utilizes transmutation products generated from nuclear reactions between high-energy protons and target materials to realize doping. Since various transmutation products exhibit different doping effects, it is promising to achieve p-type doping of gallium oxide through the Coulomb coupling effects of multiple doping elements. This study employs the Monte Carlo software FLUKA for charged particle reactions to conduct simulation analysis on transmutation doping of gallium oxide under 100 MeV proton irradiation. The results demonstrate that after 100 days of cooling post-irradiation, the activation activity decreases by approximately four orders of magnitude, and the concentration of transmutation product elements approaches stability. Analysis of the concentrations of transmutation product elements with different doping types indicates that proton irradiation transmutation can form net p-type doping. The net p-type doping concentration varies at different depths within the target material, reaching its maximum at depths of 0.6-0.9 cm, attaining 4.26$×$1014cm-3 per 1016cm-2 irradiation fluence. Compared with transmutation doping via 40 MeV proton irradiation and neutron irradiation, 100 MeV proton irradiation transmutation doping exhibits higher efficiency.

Full Text

Preamble

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Abstract

Gallium oxide, an ultra-wide bandgap semiconductor, is currently a hot research topic in the semiconductor field. However, conventional doping processes have not yet achieved p-type doping in bulk crystals, which hinders its practical applications. Proton irradiation transmutation doping utilizes transmutation products generated from nuclear reactions between high-energy protons and target materials to achieve doping. Since various transmutation products exhibit different doping effects, p-type doping of gallium oxide may be realized through the Coulomb coupling effect of multiple doping elements. This paper employs the Monte Carlo software FLUKA for charged particle reactions to simulate and analyze the transmutation doping of gallium oxide irradiated by 100 MeV protons. The results indicate that after 100 days of cooling post-irradiation, the activation activity decreases by approximately four orders of magnitude, and the concentration of transmutation product elements stabilizes. Analysis of transmutation product element concentrations with different doping types demonstrates that proton irradiation transmutation can form net p-type doping. The net p-type doping concentration varies at different depths within the target material, reaching its maximum of 4.26$\times$10^{14}$ cm^{-3} per 10^{16} cm^{-2} irradiation fluence at depths of 0.6–0.9 cm. Compared with doping via 40 MeV proton irradiation and neutron irradiation transmutation, 100 MeV proton irradiation transmutation doping exhibits higher efficiency.

Keywords: gallium oxide; doping; proton irradiation

1. Introduction

Since the birth of the first transistor at Bell Labs in the United States in the late 1940s, marking humanity's entry into the electronic age, first-generation semiconductors such as silicon and germanium have laid the foundation for the rapid development of the electronics industry centered on integrated circuits. Subsequently, second-generation semiconductors like gallium arsenide and third-generation semiconductors such as gallium nitride have flourished in power devices and other fields due to their larger bandgaps and faster electron saturation drift velocities, and have now achieved commercialization. Gallium oxide, known as the representative of "fourth-generation" semiconductors—ultra-wide bandgap semiconductor materials—offers superior performance compared to conventional wide bandgap semiconductors, with a critical breakdown field

of approximately 8 MeV/cm and a bandgap of about 4.9 eV, making it a promising semiconductor material capable of stable operation under high temperature, strong radiation, and high-frequency conditions.

In 1952, Roy et al. first reported that gallium oxide possesses five phase structures. Among these, β-phase gallium oxide is the most stable at room temperature and the most extensively studied phase for semiconductor device applications. Currently, ultra-wide bandgap semiconductor gallium oxide has found applications in solar-blind ultraviolet detectors, gas sensors, and high-power electronic devices. However, further applications of gallium oxide face the challenge of difficult p-type doping. In 2010, Liu et al. explored nitrogen doping to achieve p-type conductivity in gallium oxide nanowires, using NH_3 as the doping source and chemical vapor deposition to prepare N-doped gallium oxide nanowires, which exhibited p-type conductivity. In 2018, Adam T. Neal et al. measured the ionization energy of Mg-doped β-Ga_2O_3 to be 1.1 eV. In 2019, Su et al. achieved p-type doping of gallium oxide thin films with Mg and Zn using magnetron sputtering, determining that defects introduced by Mg_{Ga} and Zn_{Ga} created energy levels 1.00 eV and 0.79 eV above the valence band, respectively. In 2021, Jani Jesenovec et al. measured the conductivity of Zn-doped gallium oxide prepared by the Czochralski method but detected no p-type conductivity due to the deep doping levels. In 2022, Ekaterine Chikoidze et al. prepared Zn-doped gallium oxide thin films by chemical vapor deposition and, through high-temperature Hall effect measurements, determined that Zn_{Ga} introduced a doping level with an ionization energy of 0.77 eV, capable of ionizing free holes at high temperatures. Unfortunately, to date, there have been no reports of successful room-temperature p-type doping in gallium oxide single crystals. Since both n-type and p-type doping are required for applications such as bipolar device fabrication and depletion layer preparation, the difficulty in achieving p-type doping limits the development of gallium oxide materials in electronic device applications.

The reasons for the difficulty in p-type doping of gallium oxide can be summarized into three points: (1) The strong lattice structure of wide bandgap semiconductors makes conventional doping methods such as thermal diffusion and ion implantation difficult to place impurity atoms at lattice sites; (2) Theoretically, no single dopant with shallow acceptor levels has been identified; and (3) Gallium oxide with intrinsic defects exhibits weak n-type conductivity, and self-compensation effects hinder the formation of p-type doping.

Transmutation doping is a method that utilizes the transmutation of lattice atoms through nuclear reactions to achieve semiconductor doping, which can solve the problem of conventional doping methods' inability to place impurity atoms at lattice sites. Research on transmutation doping dates back to the last century. This method does not require an external doping element source but instead relies on the mechanism where excited nuclei generated by nuclear reactions decay to form new nuclei, thereby achieving doping. Transmutation doping primarily employs neutron irradiation and proton irradiation; this study focuses

on proton irradiation transmutation doping. This doping approach ensures that most doping nuclei are located at lattice sites. Due to the large range of protons in target materials, proton irradiation transmutation doping can produce more uniform impurity distributions within samples compared to conventional thermal diffusion methods. Julie V. Logan et al. used the FISPACT-II software to analyze transmutation elements in gallium oxide irradiated by 2–40 MeV protons, calculating the net p-type doping concentration and concluding that proton transmutation doping is a possible pathway to achieve p-type doping in gallium oxide.

The search for shallow-level dopants in gallium oxide is also an important research direction. Theoretically, some analyses suggest that gallium oxide has no shallow-level p-type dopant elements. Alexandros Kyrtsos et al. used density functional theory to analyze the energy levels of some metal elements in gallium oxide doping, concluding that Li, Na, Be, Mg, Ca, Cu, and Zn are all potential p-type dopants but are deep-level impurities. As research on single-element doping encounters obstacles, co-doping approaches have been anticipated with great hope. In wide bandgap semiconductors such as zinc oxide and gallium nitride, dual-element co-doping has been proven effective for achieving p-type doping due to the Coulomb coupling effect that increases carrier concentration. Liying Zhang et al. demonstrated through first-principles calculations that N-Zn co-doping in gallium oxide introduces lower ionization energy than single N doping. Cheng Tang et al. inferred based on first-principles calculations that dual-element co-doping of N with metal elements such as Sc, Ti, Cr, Mn, Fe, Cu, Ni, and Zn can reduce the bandgap of gallium oxide to achieve p-type conductivity. Among these, N-Mn and N-Cu co-doping showed the lowest calculated bandgap values of 2.51 eV and 2.12 eV, respectively. Ling Li et al. inferred based on density functional theory that N-P co-doping has lower ionization energy and smaller hole effective mass than single N or P doping, which would increase hole concentration in the semiconductor. Currently, experimental exploration of co-doping in gallium oxide remains limited, and more experimental verification is needed for the above theoretical calculations. Man Hoi Wong et al. achieved N-Mg co-doping using ion implantation, observing carrier compensation phenomena that confirmed the role of co-doping in reducing ionization energy and ionizing free holes. For proton irradiation transmutation doping, several or even more than a dozen elements are introduced into gallium oxide. While simulating the effects of doping with more than a dozen elements presents theoretical challenges, results from both theoretical calculations and experiments on dual-element co-doping suggest that multi-element co-doping is a possible approach to reduce bandgap and achieve room-temperature p-type doping in gallium oxide.

Under current process conditions, intrinsic defects are inevitably present during gallium oxide preparation. Gallium oxide crystals containing intrinsic defects (primarily oxygen vacancies) exhibit weak n-type conductivity at room temperature, which compensates for transmutation-doped p-type impurities. To avoid interference from intrinsic defects on transmutation doping results, iron-doped

gallium oxide with 0.14 wt‰ Fe will be used as the target material in actual experiments, leveraging the deep p-type impurity characteristics of iron to neutralize the n-type doping effect caused by intrinsic defects. In summary, the proton irradiation transmutation doping method theoretically can solve existing p-type doping problems and offers the possibility of achieving p-type doping in gallium oxide.

The structure of this paper is arranged as follows: Section 1 is the introduction; Section 2 introduces the simulation method for proton irradiation transmutation; Section 3 analyzes the simulation results of transmutation doping and provides recommendations for subsequent experimental work; Section 4 presents the summary and outlook.

2. Simulation Analysis Methods

The proton irradiation transmutation process was simulated using the Monte Carlo simulation software FLUKA (version 4-3.1). The irradiation conditions in the FLUKA simulation were set based on the beam capability of the high-intensity proton cyclotron at the China Institute of Atomic Energy, with proton energy specified as 100 MeV. The target material density adopted the β-phase gallium oxide density of 6.44 g/cm^3. The SRIM (2013) software was used to calculate the range of protons in gallium oxide, with results shown in Figure 1: see original paper. The range of 100 MeV protons in gallium oxide is approximately 1.71 cm. Therefore, the gallium oxide material was defined as a cylinder with a length of 1.8 cm and a radius of 1 cm, with its axis aligned with the irradiation direction, and the beam spot uniformly and completely covering the material cross-section perpendicular to the axis. Based on the data in Figure 1: see original paper, the relationship between target material depth and incident proton energy can be inferred, as shown in Figure 1: see original paper. This enables analysis of transmutation doping effects at different proton irradiation energies by examining transmutation element concentrations at various depths in gallium oxide.

During proton irradiation and subsequent cooling, lattice atoms in gallium oxide transmute into various other elemental atoms. Different elements generally exhibit different doping effects, while different isotopes of the same element show similar doping effects. Therefore, doping types are discussed by elemental classification. summarizes the doping effects and types of several elements involved in this work. It should be noted that semiconductor doping concentration and effects are not solely determined by doping elements. Lattice defects, like impurities, can disrupt the strictly periodic potential field in the crystal and introduce energy levels within the bandgap. Intrinsic and irradiation-induced defects in gallium oxide cannot be analyzed in FLUKA simulations. In this work, the n-type doping caused by intrinsic defects is considered to be neutralized by Fe elements present in the material preparation. The impact of proton irradiation defects on doping effects is more complex. A. Y. Polyakov et al. demonstrated that 10 MeV proton irradiation can increase the density of trap levels inside

gallium oxide crystals. Further analysis suggests that proton irradiation defects are primarily gallium vacancies and complex structures formed by gallium vacancies with protons, which introduce electron traps and compensate n-type doping. Kejia Wang et al. studied defects induced by higher-energy proton irradiation, confirming that under 80 MeV proton irradiation, both oxygen vacancy and gallium vacancy densities increase in gallium oxide, with oxygen vacancies increasing more significantly. These irradiation defects reduce the bandgap and shift the Fermi level upward in gallium oxide crystals. High-energy proton irradiation is more likely to displace lattice atoms from their positions, degrading crystal quality and adversely affecting device fabrication. Since the impact of such defects on doping concentration is difficult to simulate, their effects on transmutation doping are ignored in the theoretical analysis of this paper. Regardless of the doping method employed, high-temperature irradiation and post-irradiation annealing can effectively mitigate irradiation damage, similar to ion implantation doping.

The concentration of radioactive nuclides produced by irradiation varies with time according to the following relationship:

$$N(t) = N_0 e^{-\lambda t}$$

where t is time, N_0 is the radioactive nuclide concentration at $t = 0$, and $N(t)$ is the concentration at time t. λ is the decay constant, related to the nuclide half-life $T_{1/2}$ by:

$$\lambda = \frac{\ln 2}{T_{1/2}}$$

The decay processes that significantly affect the electrical properties after doping are listed below:

- $^{70}Zn \xrightarrow{n} {}^{71}Ga$
- $^{70}Ge \xrightarrow{d} {}^{71}Ga$
- $^{69}Zn \xrightarrow{n,99.6\%} {}^{69}Ga$; $^{69}Zn \xrightarrow{\beta^-,0.4\%} {}^{69}Ga$
- $^{68}Ge \xrightarrow{270.8\,d} {}^{68}Ga$; $^{68}Ge \xrightarrow{270.8\,d,100\%\,\beta^+} {}^{68}Ga$
- $^{67}Ga \xrightarrow{3.3\,d} {}^{67}Zn$; $^{67}Ga \xrightarrow{3.3\,d,100\%\,\beta^+} {}^{67}Zn$
- $^{66}Ga \xrightarrow{9.5\,h} {}^{66}Zn$; $^{66}Ga \xrightarrow{9.5\,h,100\%\,\beta^+} {}^{66}Zn$
- $^{66}Zn \xrightarrow{n} {}^{66}Cu$
- $^{65}Zn \xrightarrow{244\,d,100\%\,\beta^+} {}^{65}Cu$
- $^{64}Cu \xrightarrow{12.7\,h} {}^{64}Zn$; $^{64}Cu \xrightarrow{12.7\,h,100\%\,\beta^+} {}^{64}Zn$

- $^{64}Cu \xrightarrow{12.7\,h} {}^{64}Ni$
- $^{63}Zn \xrightarrow{38.5\,min,100\%\,\beta^+} {}^{63}Cu$
- $^{62}Cu \xrightarrow{9.7\,min} {}^{62}Ni$

3. Results and Discussion

In the FLUKA simulation, the USRBIN card was used to 统计 the activity in each region of the target material, while the RESNUCLE and DCYSCORE cards were employed to 统计 the nuclide concentrations in each region. To make the simulation results applicable to various experimental conditions, the concentrations of transmutation nuclides (elements) in the results were normalized, meaning that all subsequent calculations correspond to data at a proton irradiation fluence of 1$×10^{16}$ cm^{-2}. The simulation results show that 23 transmutation elements are produced in gallium oxide after irradiation: H, He, Li, Be, B, C, N, O, F, Ne, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, Ge, and As. At an irradiation fluence of 1$×10^{16}$ cm^{-2}, the concentrations of nuclides with significant doping effects are on the order of 10^{13}–10^{14} cm^{-3}. Accordingly, nuclides with concentrations below 10^{11} cm^{-3} have a doping concentration impact of less than one percent or even one-thousandth and are no longer analyzed in subsequent discussions. After screening, 17 elements remain in the target material: H, He, Li, Be, B, C, N, O, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, and Ge. Each of these elements exists as multiple isotopes in the target material. Approximately eighty nuclides are analytically significant, with more than half being radioactive. This paper sets the time range for target material activity analysis and transmutation element concentration analysis from the moment irradiation ends to one year after irradiation ends. Therefore, a few long-half-life nuclides, such as ^{63}Ni and ^{14}C with half-lives of hundreds or even thousands of years, are treated as stable nuclides in the discussion.

3.1 FLUKA Simulation Results of Radioactive Activity

The sample activity is extremely high immediately after proton irradiation and should be sealed in a lead container for cooling. To ensure that personnel and the environment are not affected by the radioactivity of the irradiated sample, the sample activity must be considered when processing irradiated samples. [Figure 2: see original paper] presents the simulation results of target material activity variation with cooling time, while [Figure 3: see original paper] shows the simulation results of specific activity variation with depth. In these figures, transmutation element concentrations have been converted to values corresponding to an irradiation fluence of 1$×10^{16}$ cm^{-2}.

As seen in [Figure 2: see original paper], the target material activity is on the order of 10^{11} Bq at the beginning of cooling. After one day of cooling, the activity decreases by about two orders of magnitude; after more than a month of additional storage, the activity further decreases by about two orders

of magnitude. At 100 days of cooling, the activity is reduced to below 10^7 Bq. The remaining radioactive nuclides in the material at this point are all long-half-life nuclides, such as ^{68}Ge and ^{60}Co, and the activity decreases very slowly thereafter. [Figure 3: see original paper] shows that within the depth range of 0–1.5 cm, there is no significant variation in specific activity across different regions of the target material. The specific activity in target material regions deeper than 1.7 cm decreases substantially because the proton range is 1.71 cm, and material at greater depths is minimally affected by proton irradiation.

Therefore, we select the time node of 100 days cooling in the following discussion to investigate the transmutation doping effects in gallium oxide at this point.

3.2 FLUKA Simulation Results of Transmutation Doping

The target material was divided into intervals of one millimeter in depth, and the concentrations of various elements in each interval at 100 days cooling were plotted, with points of the same element at different depths connected to obtain [Figure 4: see original paper]. Among impurity elements, the concentrations of Mn and Cr are below the lower limit in the figure and are therefore not shown.

Among the elements introduced by transmutation doping, H is the element with the highest concentration at all depths in the target material. Moreover, the concentration of H increases with depth, indicating that the depth is approaching the range of 100 MeV protons. Particularly at depths greater than 1.5 cm, the H concentration increases sharply, which would create numerous crystal defects and complex structures within the semiconductor, making it unsuitable for device fabrication. Therefore, in experiments, the thickness of the gallium oxide target material need not exceed 1.5 cm. Zn, an important element for p-type doping, shows relatively small variation in concentration distribution with thickness at depths less than 1.5 cm. In contrast, Ge, the dominant element for n-type doping, shows increasing concentration with depth. The Ge concentration at depths of 1.5–1.6 cm is approximately 4.5 times that in the 0–0.1 cm depth range, indicating that the net p-type doping concentration will decrease toward the back of the material. The concentrations of p-type doping elements such as C, N, Ni, and Cu show varying trends with increasing depth. Elements such as Co, Fe, and Mn only exhibit high concentration distributions at the front end or near the surface of the material.

To determine the depth at which the material's net p-type doping concentration is highest, this paper calculates the net p-type doping concentration at each depth based on the classification of doping types for different elements in . P-type doping elements include Li, Be, C, N, Fe, Ni, Cu, Zn, and Mn; n-type doping elements include Ge and Cr. The net p-type doping concentration is obtained by subtracting the sum of n-type doping element concentrations from the sum of p-type doping element concentrations. Co and B are not included in the 统计. The concentration of Co at the front end of the target material is one order of magnitude lower than that of major impurity elements such as

Ge and Zn, and in the middle region, its concentration is less than one percent of major impurity element concentrations. Therefore, excluding Co from the concentration calculation does not affect the net p-type doping concentration 统计. Although the concentration of B can reach the order of 10^{13} cm^{-3}, its concentration is essentially the same across all regions except for a significant decrease at the back of the material (depth > 1.4 cm). Thus, excluding B from the concentration calculation also has minimal impact on the net p-type doping concentration 统计. [Figure 5: see original paper] plots the net p-type doping concentration at different depths per unit irradiation fluence at 100 days cooling; data for 50 days cooling are also provided as a reference for doping effects at shorter cooling times.

Two conclusions can be drawn from the above data: First, at all depths, samples with longer cooling times exhibit higher net p-type doping concentrations. Second, at 100 days cooling, samples in the depth region of 0.6–0.9 cm show the highest net p-type doping concentration, reaching 4.26$×10^{14}$ cm^{-3} per 1$×10^{16}$ cm^{-2} irradiation fluence. Combining this with the proton energy data at various depths from Figure 1: see original paper, we can infer that the highest net p-type doping concentration in gallium oxide is achieved under 70$±$5 MeV proton irradiation conditions. The above conclusions do not consider deep-level impurity ionization ratios or multi-element co-doping effects, as these factors are difficult to simulate accurately and must be determined based on actual experiments.

3.3 Comparative Analysis of Transmutation Doping Simulation Results

First, the FLUKA simulation results are compared with literature [26] proton irradiation simulation results to verify the reliability of this simulation. Julie V. Logan et al. used the FISPACT-II software in 2020 to analyze transmutation element concentrations in gallium oxide irradiated by 2–40 MeV protons and calculated net p-type doping concentrations [26]. They set up six simulation conditions: proton energies of 2, 5, 10, 20, 30, and 40 MeV, all with an irradiation fluence of 2.25$×10^{17}$ cm^{-2}. The simulation results showed that 40 MeV protons achieved the best p-type transmutation doping effect, with a net p-type doping concentration of 8.1$×10^{15}$ cm^{-3}.

We selected the maximum energy of 40 MeV from the six groups for comparison. Since the transmutation effects of different proton energies in this simulation were obtained by acquiring element concentrations at different depths in the target material, and the minimum depth increment in the simulation is 1 mm, the simulation data represent the average transmutation doping effect of the highest and lowest energy protons within a 1 mm depth range at a given location. As shown in Figure 1: see original paper, the larger the energy, the smaller the proton energy range covered by a 1 mm thick material, and thus the smaller the comparison error. For example, 2 MeV and 40 MeV protons are located at depths of 1.7–1.8 cm and 1.3–1.4 cm, respectively, in the FLUKA simulation.

The depth range of 1.7–1.8 cm covers a proton energy range of approximately 0–7 MeV, deviating from the comparison value of 2 MeV by several times; the depth range of 1.3–1.4 cm covers a proton energy range of approximately 38–41 MeV, with a maximum deviation of only 5% from the comparison value of 40 MeV.

The time-dependent concentrations of major transmutation elements in gallium oxide at a depth of 1.3–1.4 cm are shown in [Figure 6: see original paper]. The FLUKA simulation results show that at 100 days cooling, the H concentration is the highest, approaching 1$×10^{15}$ cm^{-3}; among metal elements, Zn has the highest concentration at about 2$×10^{14}$ cm^{-3}; other metal elements in descending order of concentration are Ge, Cu, and Ni, all at the order of 10^{13} cm^{-3}; among non-metal elements, C and N have relatively high concentrations, while Be has a lower concentration. In literature [26], under the same cooling duration after 40 MeV proton irradiation, the H concentration exceeds 10^{16} cm^{-3}; among metal elements, Zn has the highest concentration in the range of 10^{15}–10^{16} cm^{-3}; other metal elements in descending order of concentration are Ge, Cu, and Ni, with concentrations ranging from the order of 10^{15} cm^{-3} to nearly 1$×10^{14}$ cm^{-3}. Among other non-metal elements, C and N have relatively high concentrations, while Co, Fe, and F have concentrations lower than Be. In the FLUKA simulation, the concentrations of Co, Fe, and F at this depth are below 10^{11} cm^{-3} and were ignored in the [Figure 6: see original paper] 统计. To compare the two simulation results, the element concentration values in literature [26] were normalized to a fluence of 1$×10^{16}$ cm^{-2}. The calculated results for the six elements with the highest concentrations from both simulations are summarized in . The data in literature [26] were obtained from figures, and reading errors exist when 引用 numerical values.

In addition to single-element concentration calculations, literature [26] also calculated the net p-type doping concentration in gallium oxide after 40 MeV proton irradiation and 50 days cooling. After converting the irradiation fluence to 1$×10^{16}$ cm^{-2}, the net p-type doping concentration was 3.6$×10^{14}$ cm^{-3} [26], approximately 85% of the net p-type doping concentration produced by 75 MeV proton irradiation in this work. The calculation result under the same conditions in this simulation is 3.79$×10^{14}$ cm^{-3}, with a deviation of about 5% from the literature value. The deviation arises from differences in simulation software models, truncation errors in doping concentration calculations, and deviations between the proton energy range at depth 1.3–1.4 cm and monoenergetic 40 MeV. Based on the above analysis, for net p-type doping concentration calculations, the results of this simulation are consistent with those of literature [26] within a 5% error range.

Second, the FLUKA simulation results are compared with neutron irradiation simulation results from literature [26] to demonstrate the superiority of proton simulation transmutation doping effects. Julie V. Logan et al. used the FISPACT-II software to simulate the effects of 12 different neutron energy spectra as neutron sources on gallium oxide transmutation doping [26]. All neutron

sources had an irradiation fluence of 3.6$×10^{17}$ cm^{-2}. The simulation results showed that the concentrations and types of elements introduced by transmutation doping are closely related to the neutron source emission spectrum. Among the 12 neutron sources, only ^{252}Cf and JAEA-FNS sources showed potential for achieving p-type doping in gallium oxide. The JAEA-FNS source produced the maximum net p-type doping concentration from transmutation doping, with a net p-type doping concentration in gallium oxide of approximately 2.39$×10^{14}$ cm^{-2} when normalized to a fluence of 1$×10^{16}$ cm^{-2}. Compared with the simulation results in this work, the net p-type doping concentration produced by neutron irradiation transmutation doping is lower, approximately 56% of that produced by 75 MeV proton irradiation.

4. Summary and Outlook

Currently, the application of gallium oxide faces challenges due to the difficulty in achieving p-type doping. Proton transmutation doping offers the advantage of placing doping atoms at lattice sites and enables multi-element co-doping, providing a possible pathway to realize p-type doping in gallium oxide. This paper uses the FLUKA software to simulate the process of 100 MeV proton irradiation of gallium oxide and presents simulation data for target material activity and transmutation element concentrations per unit irradiation fluence. The results show that after 100 days of cooling, the specific activity of the target material will be below 10^7 Bq/cm^3. In the depth region of 0.6–0.9 cm of the target material, the net p-type doping concentration in gallium oxide is maximized: at a proton fluence of 1$×10^{16}$ cm^{-2}, gallium oxide cooled for 100 days can achieve a net p-type doping concentration of 3.79$×10^{14}$ cm^{-3}. Based on the relationship between range and proton energy, we infer that proton irradiation at an energy of 70$±$5 MeV yields the best transmutation p-type doping effect. The transmutation doping effect at this energy is also superior to other proton irradiation energies and neutron irradiation transmutation doping effects calculated in literature [26]. Comparison of this simulation with literature [26] shows that the net p-type doping concentration calculation results are consistent within a 5% error range. Due to practical factors such as incomplete ionization of doping impurities, presence of irradiation defects in gallium oxide, and incomplete compensation between intrinsic defects and iron atom doping under actual working conditions, the actual net p-type doping concentration achieved by proton irradiation of gallium oxide may be lower than the simulation results. Future work will focus on experimental verification of the actual doping effects.

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