

PHYSICAL AND TECHNOLOGICAL ASPECTS OF THE METHOD OF NEUTRON TRANSMUTATION DOPING OF SEMICONDUCTORS AND PROSPECTS OF ITS APPLICATION IN UKRAINE

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Neutron transmutation doping (NTD) is a state-of-the-art alternative to traditional doping methods, such as thermal diffusion and ion implantation. Traditional methods have restrictions, in particular, a problem of uneven distribution of impurities along the ingot due to a high segregation coefficient. NTD overcomes these drawbacks ensuring uniform formation of impurity throughout the entire volume. Physical basis of the method is the absorption of thermal neutrons with ³⁰Si nuclei, which leads to creation of intermediate radioactive isotope ³¹Si. Then this isotope decays, turning into stable doping element-phosphorus (³¹P). The probability of this process is characterized by the reaction cutoff cross-section which is the biggest for the thermal neutrons. Modeling of isotope accumulation kinetics shows that phosphorus concentration increases linearly with irradiation time allowing precise control of the doping level.

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INTRODUCTION

The background of modern semiconductor electronics is single-crystal silicon of exceptionally high purity. Achieving and maintaining such purity is one of the most difficult technological tasks solved by leading global manufacturers, such as Wacker Chemie (Germany) and Shin Etsu (Japan) [1]. However, for the creation of semiconductor devices, pure silicon is only the starting raw material. The key stage is doping – the controlled introduction of impurities to create n- or p-type conductivity. The growing need to produce powerful and high-voltage devices, which are the basis for green energy, electric transport, and stable power grids, places increasingly stringent requirements on the uniformity and accuracy of doping [2].

Traditional doping methods, such as thermal diffusion and ion implantation, face fundamental limitations in solving these problems. The problem of uniformity and accuracy of doping is rigorous in creating n-type silicon devices, where phosphorus is the dopant [3, 4]. Traditional methods here face fundamental limitations. Phosphorus has a significant segregation coefficient in silicon ($k \approx 0.35$), which, when growing crystals by the Czochralski method or zone melting, leads to a strong non-uniformity of the dopant distribution along the ingot [5, 6]. Diffusion methods at high temperatures do not allow for uniform distribution in large volumes and create uncontrolled concentration profiles. Although ion implantation is a low-temperature and precise method, it remains a near-surface technology, which also causes significant radiation damage to the crystal lattice. Further annealing to eliminate these defects is not always 100% effective and can lead to additional diffusion and redistribution of the impurity [7].

An alternative to these methods is the neutron transmutation doping (NTD) method, which is based on converting the stable isotope ³⁰Si into the doping impurity ³¹P using a flux of thermal neutrons. This technolo-

gy is not new and is actively used worldwide in dozens of research nuclear reactors with different neutron flux parameters, allowing for flexible selection of irradiation modes [8]. Due to the high penetrating power of neutrons, NTD allows achieving extremely high doping homogeneity (heterogeneity < 1...2%), which is unattainable for other methods.

This research aims to provide a comprehensive overview of the NTD method, including its fundamental physical background, technological aspects, analysis of advantages and disadvantages, and consideration of the prospects for its application in the scientific and technical scope of applications in Ukraine.

1. PHYSICAL FOUNDATIONS OF NTL

The NTP method is based on the fundamental processes of neutron interaction with matter and subsequent nuclear transformations. To understand the process, it is necessary to consider three key aspects: the energy spectrum of neutrons used for irradiation, the cross sections of the corresponding nuclear reactions, and the kinetics of the formed isotope accumulation.

1.1. REACTOR NEUTRON SPECTRUM

The NTP is based on using a neutron flux generated in a nuclear reactor or any other intense neutron source. However, only thermal neutrons are used for doping. Due to numerous collisions with moderator atoms (for example, water, graphite, or beryllium), these are slowed (thermalized) neutrons that have reached a state of thermal equilibrium with the medium.

The Maxwell distribution describes the energy spectrum of such neutrons, which for a typical moderator temperature of 300 K has a maximum in the region of very low energies – about 0.025 eV. The real spectrum of neutrons in the experimental reactor channel also includes the so-called “epithermal tail”, where the flux decreases approximately according to the law $1/E$ (Fig. 1). It is in the low-energy thermal region that the

cross sections of the radiative neutron capture reactions (n,γ) for many isotopes are the largest [9].

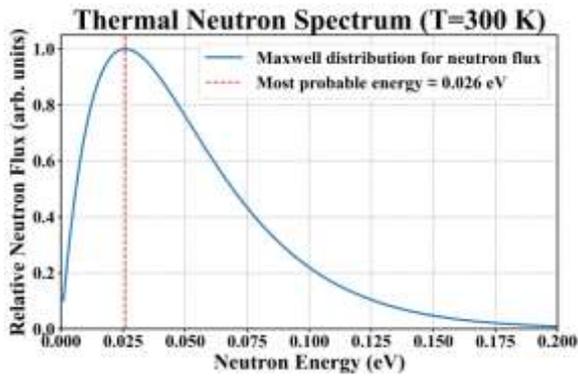


Fig. 1. Representative Maxwell spectrum of neutrons in the experimental channel of a research reactor, showing the thermal Maxwell peak and the epithermal region [9]

1.2. NUCLEAR REACTIONS CROSS-SECTIONS

The probability of a neutron interacting with a nucleus is characterized by a quantity called the reaction cross section (σ). For the paper, the σ_{theory} cross sections of the radiative neutron capture reactions (n,γ) for three stable silicon isotopes were calculated with using the TALYS code (version 1.96) in the default mode [10]. The calculations were performed on an energy grid that covers the range from 10^{-9} to 2 MeV to describe the thermal and qualitatively resonant regions in detail. The relative composition is of ^{28}Si – 92.2% ($4.61 \cdot 10^{22}$ at./cm³); ^{29}Si – 4.67% ($2.34 \cdot 10^{21}$ at./cm³); ^{30}Si – 3.1% ($1.55 \cdot 10^{21}$ at./cm³).

Fig. 2 shows the dependence of the calculated cross sections on the neutron energy. It is worth noting (to note) that although the energy grid used describes the general behavior of the cross sections well, its step may be insufficient to reproduce narrow resonance peaks in the epithermal region accurately. However, the thermal energy region is the key to the NTD process. In the thermal energy region (0.0253 eV), the calculated cross-section values agree well with the known experimental data, confirming the model's correctness in this key range for NTD – the comparison of the data presented in Table. As can be seen from the data, although all three isotopes interact with thermal neutrons, their cross sections are of the same order of magnitude. The decisive factor for the doping process is not the cross-section value, but the properties of the reaction product. Only the reaction on ^{30}Si nuclei leads to the formation of the intermediate radioactive isotope ^{31}Si and, as a result, to the appearance of a stable phosphorus doping impurity.

The formed radioactive isotope ^{31}Si is unstable and undergoes beta decay (β^- -decay), transforming into a stable isotope of phosphorus (^{31}P), and the primary alloying element in silicon. The key properties of the ^{31}Si nucleus that determine the kinetics of the process are [12]:

Half-life ($T_{1/2}$): is approximately 2.62 h (157.3 min).
Beta particle energy (E_β): the maximum energy with which electrons are emitted is 1.492 MeV. This half-life is short enough that most radioactive isotopes decay within 2–3 days after irradiation. Therefore, the material

requires some time to "cool down" before further processing.

Comparison of calculated and experimental cross sections for thermal neutrons ($E_n=0.0253$ eV)

Nuclear reaction	σ_{theory} , barn	σ_{exp} [11], barn
$^{28}\text{Si}(n,\gamma)^{29}\text{Si}$	≈ 0.16	0.16 ± 0.02
$^{29}\text{Si}(n,\gamma)^{30}\text{Si}$	≈ 0.11	0.107 ± 0.003
$^{30}\text{Si}(n,\gamma)^{31}\text{Si}$	≈ 0.10	0.108 ± 0.002

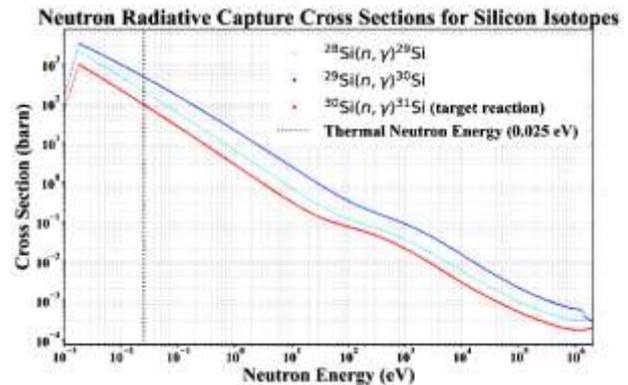


Fig. 2. General behavior of neutron capture cross sections for stable silicon isotopes as a function of neutron energy, calculated using the TALYS code

2. KINETICS OF ACCUMULATION AND DECAY OF ISOTOPES

For a detailed analysis of the change in the isotopic composition of silicon during irradiation, a system of differential equations was solved, describing the kinetics for each key isotope. The calculations were done for a thermal neutron flux, $F_n=5 \cdot 10^{13}$ neutr./cm²·s). The system of equations has the following form (1):

$$\begin{cases} \frac{dN_{28}}{dt} = -N_{28} - \sigma_{28} F_n \\ \frac{dN_{29}}{dt} = N_{28} \sigma_{28} F_n - N_{29} \sigma_{29} \\ \frac{dN_{30}}{dt} = N_{29} \sigma_{29} F_n - N_{30} \sigma_{30} F_n \\ \frac{dN_{31}}{dt} = N_{30} \sigma_{30} F_n - \lambda_{31} N_{31}(t) \end{cases} \quad (1)$$

where σ_i is the reaction cross section (n,γ) for the corresponding silicon isotope, and λ_{31} is the decay constant of ^{31}Si .

The solution of the system for impurity and intermediate isotopes is presented in Fig. 3. Analysis of the graph shows that the concentration of the doping impurity ^{31}P increases almost linearly with time, which is key for precise control of the doping level. The model confirms that an irradiation time of several to tens of hours is required to achieve the target levels typical for power electronics ($10^{13} \dots 10^{15}$ at./cm³). At the same time, the concentration of the radioactive isotope ^{31}Si quickly reaches an equilibrium state, when the rate of its formation is equal to the rate of its decay.

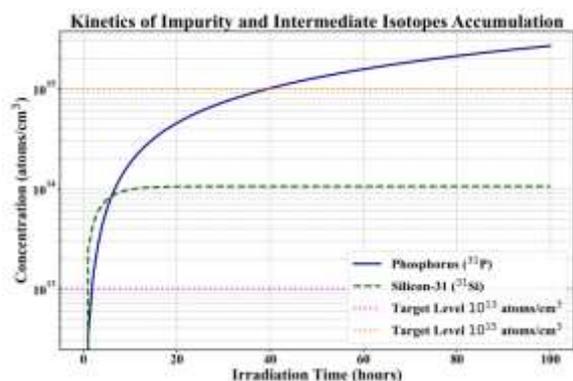


Fig. 3. Dynamics of impurity accumulation and intermediate isotopes during NTL of silicon. Horizontal lines show target doping levels

During the 100-h irradiation period, the isotopic composition of the silicon crystal undergoes significant transformation. The concentration of the primary isotope, ^{28}Si , slightly decreases from $4.61 \cdot 10^{22}$ to $4.57 \cdot 10^{22}$ at./cm 3 as it transmutes via neutron capture. Consequently, the concentrations of ^{29}Si and ^{30}Si increase modestly, reaching $2.37 \cdot 10^{21}$ and $1.56 \cdot 10^{21}$ at./cm 3 , respectively. The key intermediate isotope, ^{31}Si , which is unstable, accumulates from zero to a concentration of $7 \cdot 10^{14}$ at./cm 3 . Its subsequent beta decay leads to the formation of the desired dopant, ^{31}P , which reaches a final concentration of $1.21 \cdot 10^{16}$ at./cm 3 , achieving the goal of the transmutation process.

3. TECHNOLOGY AND THE ADVANTAGES OF NTL

The industrial process of the NTD is multi-stage and requires strict adherence to technology. It includes the following key stages:

1. Preparation and Measurement. The starting material should be high-purity single-crystal silicon ingots, usually grown by zone melting. Before irradiation, the initial resistivity is carefully measured to calculate the required neutron dose (fluence) accurately.

2. Irradiation in a nuclear reactor. Ingots are packed in special containers (usually made of aluminum) and placed in one of the vertical experimental channels of a nuclear reactor. They are irradiated with a flux of thermal neutrons during a certain time (from several hours to tens of hours). To achieve maximum uniformity of fluence throughout the volume, containers with ingots are usually rotated during irradiation.

3. Radiation “cooling”. After irradiation, silicon ingots become radioactive due to the formation of the intermediate isotope ^{31}Si and the activation of impurities. They are moved to a special storage (a wind pool or “hot chamber”) for a specific time, sufficient for the decay of short-lived radionuclides to a safe level (usually a few days).

4. Thermal annealing. Irradiation, especially the accompanying component of fast neutrons and gamma rays, creates radiation defects (vacancies, interstitial atoms) in the crystal lattice, significantly worsening the material's electrical properties. To eliminate them and electrically activate the formed phosphorus atoms (so that they occupy the correct positions in the lattice and become donors), high-temperature annealing of ingots is

carried out (usually at temperatures of 800...900 °C).

5. Output control. After annealing, a final measurement of the resistivity across the entire alloy is completed to confirm that the specified parameters have been achieved and that the alloying is highly uniform.

Despite its technological complexity, the NTD method has several unique advantages that make it indispensable for specific applications:

Exceptional uniformity. The high penetration of thermal neutrons ensures uniform formation of impurity atoms throughout the entire volume of even enormous ingots (diameter 100...200 mm, height of 1 m). The non-uniformity of resistivity in NTD silicon usually does not exceed 1...2%, while this figure can reach 20...30% for traditional methods.

High accuracy and reproducibility. The final phosphorus concentration is directly proportional to the neutron fluence (flux yield per time), which can be controlled with very high accuracy. This allows for the reliable production of material with predetermined electrical properties.

Purity of the process. Doping occurs without introducing any foreign chemical impurities, which allows for maintaining the initial high purity of silicon.

Raw material quality control. Thermal neutron irradiation allows, simultaneously with doping, to use neutron activation analysis (NAA), one of the most sensitive methods for determining the elemental composition. Analyzing the gamma spectra of activated impurities makes it possible to control the purity of the starting raw material with high accuracy [13]. At the final stage of quality control of semiconductor silicon, it is possible to involve nuclear-physical methods for determining impurity content and spatial distribution when using small-sized charged particle accelerators [14, 15].

4. APPLICATION PROSPECTS IN UKRAINE

Consideration of the possibility of implementing NTD technology in Ukraine is a pressing task that can contribute to the development of high-tech industries and strengthen the scientific and industrial potential of the state. Analysis of prospects requires consideration of both existing prerequisites and potential challenges.

4.1. PREREQUISITES

A necessary but not sufficient prerequisite for the implementation of NTD is the availability of powerful thermal neutron sources. Typically, the method is used in the core of research reactors where thermal neutrons with a high neutron flux can be used, and the production volume is small. Typically, such research reactors operate with crystals measuring 50 cm in length and 20 cm in diameter. For example, such as Orphée at Saclay, France, and Forschungsreaktor München II (better known as FRM II at Garching, Germany [16]).

Thus, one of the key prerequisites for implementing the NTD method in Ukraine is the operation of the VVR-M research reactor at the Institute for Nuclear Research of the National Academy of Sciences of Ukraine [17]. Its characteristics, in particular the presence of vertical experimental channels with a high flux of thermal neutrons (up to 10^{14} n/(cm 2 ·s)), are technical-

ly suitable for the irradiation of silicon ingots to develop the technological foundations of NTD.

Even though the VVR-M operation term has been extended, the global tendency indicates that the number of research reactors will be decreased yearly. Thus, in June 2025, there were 847 research reactors in 72 countries around the world, of which 227 in 54 countries were in operation, 23 were under construction, and 73 reactors were shut down [18]. It is estimated that the number of research reactors worldwide will be decreased over the next 20 years.

This suggests that researchers must find other ways to create powerful neutron sources.

One such way is Subcritical neutron sources, which are controlled by particle accelerators [19]. Currently, several projects of such facilities are being designed and built in the world [20], one of which is the Nuclear Subcritical Facility "Neutron Source" of the NSC KIPT [21].

The facility has passed the physical start-up stage [22, 23]. After reaching the design power, it will provide a neutron flux of $3 \cdot 10^{14}$ n/s directly near the neutron-generating target with an amplification factor of 50 ($k_{\text{eff}} = 0.98$). The Nuclear Subcritical Facility "Neutron Source" of the NSC KIPT has the necessary infrastructure (including a storage pool) and hot chambers, which will allow for the full cycle of implementing and improving the NTD technology for domestic production needs.

Another possibility for the implementation of NTD technology is provided by the so-called compact neutron sources (CANS) based on accelerators [24, 25]. CANS are neutron sources that use accelerators, but do not use the spallation reaction to generate neutrons. Instead, such devices use the interaction of relativistic particles with the material of the neutron-generating target made of heavy metals (tungsten, uranium, etc.). The most common types of accelerators used in CANS are cyclotrons, electrostatic, and RF accelerators of particles such as electrons and protons.

Currently, based on the progress in creating of powerful accelerators in the world, the experience of NSC KIPT in the development and use of accelerators [26–29] and neutron-generating targets [30], and the availability of the necessary infrastructure, it can be stated that NSC KIPT is capable of creating an accelerator complex for implementing the NTD method.

Scientific and technical base. In Ukraine, based on the Institute of Nuclear Research of the National Academy of Sciences of Ukraine, the National Scientific Center "Kharkiv Institute of Physics and Technology", and leading universities, a powerful scientific school in the field of solid-state radiation physics, semiconductor physics, and nuclear physics has historically developed. There is a human resource potential capable of providing scientific support for the development and improvement of NTD technology.

Potential market. There is both a domestic and external market for high-quality NTD silicon. Potential consumers may include Ukrainian enterprises developing power electronics and specialized detectors, as well as foreign companies requiring materials with unique properties.

4.2 TECHNOLOGICAL AND ORGANIZATIONAL CHALLENGES

Infrastructure modernization. For industrial production, it is necessary to develop technological systems at neutron sources for precise positioning and uniform rotation of silicon ingots during irradiation, as well as install online neutron fluence monitoring systems for precise dose control.

Developing a complete technological chain. The implementation of the method requires the organization of the entire cycle: from the purchase and incoming control of high-purity ingots to the creation of infrastructure for safe transportation, temporary storage ("cooling") of activated materials, and their subsequent high-temperature annealing.

Certification and quality control. To enter the international market, it is necessary to have a quality control laboratory equipped to measure the electrophysical parameters of NTD silicon at the world level. The products must undergo appropriate international certification. Thus, the implementation of the NTD project in Ukraine is a complex but strategically important task that requires public or private investments and close cooperation between scientific institutions and industry.

CONCLUSIONS

Based on the analysis of the physical foundations, technological aspects, and prospects for the application of the NTD technique, the following conclusions can be drawn:

1. The NTD method is a unique technology that allows achieving the highest degree of uniformity and accuracy of doping of semiconductor materials, in particular silicon. It is based on the process of converting the crystal's own atoms into atoms of an alloying impurity under the action of a thermal neutron flux.

2. The key advantages of the method, unattainable for traditional diffusion and ion implantation technologies, are the extreme homogeneity of the impurity distribution over the volume of large ingots, high reproducibility and accuracy of dopant concentration control, as well as the purity of the process.

3. Ukraine has the necessary scientific and technical prerequisites for the implementation of NTD technology, namely: an operating VVR-M research nuclear reactor with appropriate neutron field characteristics and a powerful scientific school in radiation physics and materials science, a powerful neutron source of the NSC KIPT based on a subcritical assembly driven by an electron accelerator, which is at the commissioning stage, the necessary infrastructure for the implementation of NTD technology and the scientific and technical potential for the development and implementation of an accelerator complex optimized for NTD technology.

4. The implementation of the NTD project in Ukraine is a complex but strategically important task. It requires the modernization of the reactor infrastructure, the creation of a complete technological chain (including annealing and control stages), and product certification. Successfully solving these tasks may allow Ukraine to occupy its own niche in the world market of high-tech materials for power electronics.

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ФІЗИЧНІ ТА ТЕХНОЛОГІЧНІ АСПЕКТИ МЕТОДУ НЕЙТРОННОГО ТРАНСМУТАЦІЙНОГО ЛЕГУВАННЯ НАПІВПРОВІДНИКІВ ТА ПЕРСПЕКТИВИ ЙОГО ЗАСТОСУВАННЯ В УКРАЇНІ

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Метод нейтронного трансмутаційного легування (НТЛ) є сучасною альтернативою традиційним методам легування, таким як термічна дифузія та іонна імплантація. Традиційні методи мають обмеження, зокрема проблему нерівномірності розподілу домішки вздовж злитка через високий коефіцієнт сегрегації. НТЛ долає ці недоліки, забезпечуючи рівномірне утворення домішки по всьому об'єму. Фізична основа методу полягає у взаємодії теплових нейтронів з ядрами ^{30}Si , що призводить до утворення проміжного радіоактивного ізотопу ^{31}Si . Цей ізотоп потім розпадається, перетворюючись на стабільний легуючий елемент – фосфор (^{31}P). Імовірність такої взаємодії характеризується перерізом реакції, який для теплових нейтронів є найбільшим. Моделювання кінетики накопичення ізотопів показує, що концентрація фосфору зростає лінійно з часом опромінення, що дозволяє точно контролювати рівень легування.