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INFLUENCE OF GAMMA RAYS ON ELECTROPHYSICAL PROPERTIES OF SILICON DOPED WITH PALLADIUM ATOMS

Sh.B. Utamuradova[1], **Sh.Kh. Daliev**[1], **D.A. Rakhmanov**[1*], **I.Kh. Khamidjanov**[1], **A.S. Doroshkevich**[2,3], **Zh.V. Mezentseva**[2], **A. Tatarinova**[2]

[1]Institute of Semiconductor Physics and Microelectronics at the National University of Uzbekistan, 20 Yangi Almazar st., Tashkent, 100057, Uzbekistan
[2]Joint Institute for Nuclear Research, 141980, Dubna, Russia
[3]Dubna State University, Dubna, Universitetskaya str., 19, Russian Federation

Abstract. In this paper, we study the effect of gamma radiation (dose 10^7 rad) on the electrophysical properties of single crystals of n-type silicon (n-Si) doped with palladium atoms at different temperatures using the impedance spectroscopy method. It is determined that the electrical resistance of the original silicon will sharply increase several times as a result of irradiation due to the creation of radiation defects. In samples doped with Pd, γ-irradiation, on the contrary, reduces the resistance. This may be due to the destruction of existing traps, their transition to a neutral state or a shift in energy levels, reducing their activity. Using an electron microscope, the elemental composition was determined and micrographs were obtained on the surface of the original doped silicon samples.

Keywords: silicon, palladium, doping, diffusion, gamma radiation, impedance spectroscopy.
[*]**Corresponding author:** D.A. Rakhmanov, Institute of Semiconductor Physics and Microelectronics at the National University of Uzbekistan 20 Yangi Almazar st., Tashkent, 100057, Uzbekistan, Tel: +998974651416, e-mail: *dilmurod-1991@bk.ru*
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1 Introduction

In the context of the rapid advancement of modern microelectronics and semiconductor technology, an important scientific and technical challenge remains the comprehensive investigation of novel materials and their properties. Among them, silicon-based structures represent the most extensively utilized semiconductor materials in industry. Through doping with various elements, the electronic and electrophysical characteristics of silicon can be deliberately modified to achieve the desired properties. One dopant that has attracted particular attention is palladium (Pd), as its incorporation into the silicon crystal lattice significantly alters the electronic states and defect structure of the material (Utamuradova et al., 2006; 2023; Ahmadova & Jabarov, 2023).

Palladium can act as a deep acceptor level in silicon, which directly affects the electrical conductivity of the semiconductor. One of the features of such structures is their sensitivity to external influences, including ionizing radiation, such as gamma rays. Gamma radiation can cause the appearance of new defects in the crystal lattice of silicon or activate existing ones, thereby changing the electrophysical parameters of the material. Therefore, the study of the behavior of palladium-doped silicon under the influence of gamma radiation is of both theoretical and practical interest (Nabiyeva et al., 2024; Utamuradova et al., 2024).

Previous studies have mainly focused on the influence of traditional alloying elements (such as phosphorus, boron) and their radiation resistance, while the influence of heavy metal atoms such as palladium has been poorly studied. In particular, there is little information on what new defect states arise in such structures under the influence of high-energy radiation (gamma rays) and how these states affect the electronic properties. The present study aims to fill this gap (Azamatov et al., 2023; Utamuradova et al., 2023; 2024; Abdurakhmanov et al., 1998).

The aim of this work is to study the influence of gamma radiation on the electrical properties of n-Si and n-Si<Pd> single crystals using impedance spectroscopy.

This article examines how the electrophysical properties of palladium-doped silicon structures change under the influence of gamma radiation. The results of the study open up new possibilities for increasing the radiation resistance of semiconductor materials and developing modified structures suitable for use in space technologies and devices operating under nuclear radiation conditions.

2 Experimental part

The objects of study were n-type silicon wafers with dimensions of $1 \times 6 \times 13$ mm and a specific resistivity of 40Ω·cm (KEF-40). The wafers were cut from silicon ingots grown by the Czochralski method. The oxygen concentration in the initial n-Si samples was 6.2×10^{17}cm^{-3}. Doping with palladium was carried out by the diffusion method, with palladium atoms deposited on the silicon surface in evacuated quartz ampoules at temperatures of $1100 - 1200$°C for 3–5 hours.

After cleaning, both the original and doped samples were irradiated with gamma rays of 1.25 MeV energy at a total dose of 1×10^7 rad (dose rate: 75.6 R/s) at a temperature of 30 ± 2°C. The irradiation was carried out using a ^{60}Co source at the Institute of Nuclear Physics, Academy of Sciences of the Republic of Uzbekistan (INP AN RUz).

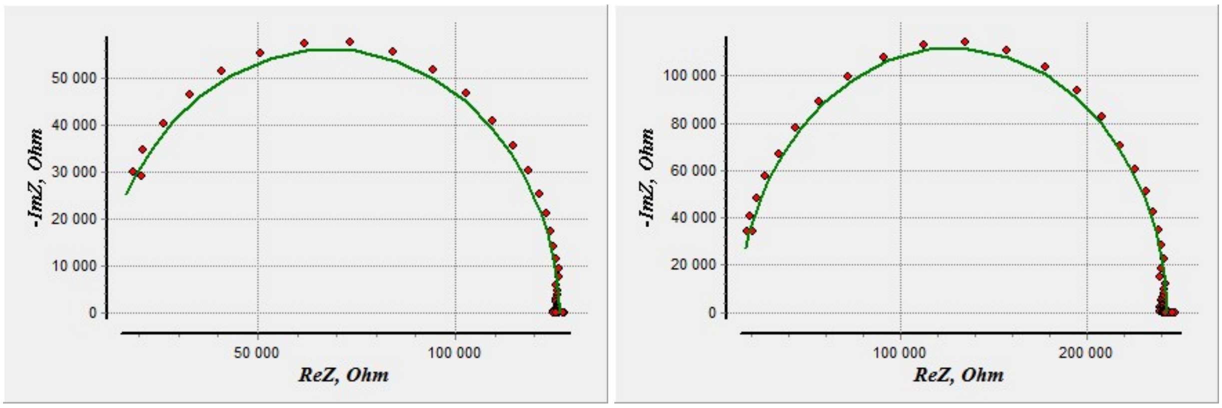
The electrical properties of the silicon samples were studied at room temperature (300 K) using an Ellins P-45X impedance meter in the frequency range of 1 Hz to 0.5 MHz with an excitation signal voltage of 10 mV. The measurements were performed in air (without vacuum), and silver pressure contacts were used as external electrodes. Micrographs of the samples were obtained using a scanning electron microscope (SEM). The elemental composition of the silicon samples was analyzed by energy-dispersive spectroscopy (EDS) in combination with electron microscopy.

3 Results and their discussion

Fig. 1 shows the impedance hodographs of an undoped n-Si sample before and after irradiation (hereinafter referred to as "initial"). The impedance spectrum (Fig. 1,a) is in the resistance range of 18.5kΩ$\div$127.7kΩ and has a semicircle shape, which is typical for the electrode reaction. The section of the diagram in the range from $0 \div 18.5$kΩ along the X axis corresponds to the electrolyte resistance (Emelyanova et al., 2017).

The hodographs of the irradiated sample (Fig. 1,b) have a similar shape and are in the range of 17 kΩ $\div$ 247.3 kΩ. It can be seen that irradiation in this case led to an increase (almost 2 times) in the resistance of the sample, presumably due to the formation of radiation defects in the crystal lattice (Vladikova et al., 2011; Utamuradova et al., 2023). The most significant factor of radiation exposure is radiation defect formation, in particular, the formation of vacancies. Such defects primarily include oxygen vacancies O_i - vacancy V (A-center), divacancies (V-V) and the phosphorus complex P_S (at the site) - vacancy V (E-center) (Kozlov & Kozlovskiy, 2001).

The hodographs of the studied samples can be adequately described by a single equivalent electrical circuit (Fig. 2). It should be emphasized that the scatter of points in the low-frequency region is significantly smaller compared to the unirradiated sample. This indicates a relatively high homogeneity of the conductivity channels after irradiation, which is consistent with the previously reported data (Nijen et al., 2023; Tetelbaum et al., 2022).



(a) (b)

Figure 1: Hodographs of the initial n-Si sample: before (a) and after (b) irradiation. The green line is the best fit curve

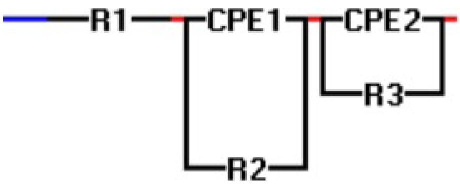


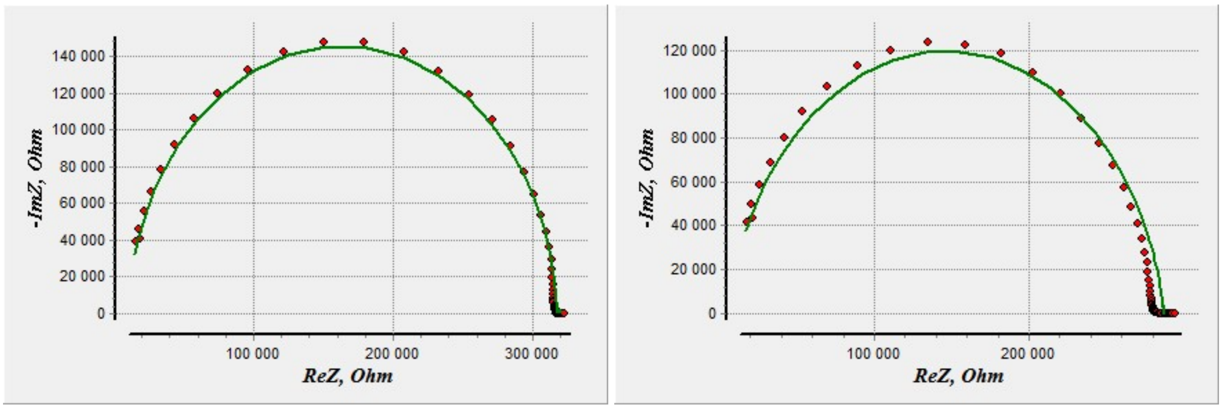
Figure 2: Equivalent electrical circuit, where R is the active resistance; CPE is a constant phase element.

We also determined equivalent electric circuit parameters using software "EIS Spectrum Analyser". Below in Table 1 are the values of the equivalent electric circuit parameters. These values also undergo changes after doping and irradiation (Table 1).

Table 1: Summary table for the equivalent electric circuit parameters

Parameters	**n-Si**	**n-Si (irradiated)**	**n-Si<Pd> (1100 ^{0}C)**	**n-Si<Pd> (1100 ^{0}C, irradiated)**	**n-Si<Pd> (1200 ^{0}C)**	**n-Si<Pd> (1200 ^{0}C, irradiated)**
R_1	10051	12572	10292	5687.6	7366.1	6990.5
R_2	58264	115240	3038.4	140580	63778	38483
R_3	58264	115240	307270	140580	65747	38483
P_1	3.3652×10^{-11}	2.9791×10^{-11}	6.4467×10^{-6}	7.1088×10^{-11}	3.4247×10^{-11}	5.7381×10^{-11}
n_1	0.97608	0.98159	0.93965	0.89824	0.93541	0.92835
P_2	3.3652×10^{-11}	2.9791×10^{-11}	1.5622×10^{-11}	7.1088×10^{-11}	2.1029×10^{-11}	5.7381×10^{-11}
n_2	0.97608	0.98159	0.9674	0.89824	1.0098	0.92835

Interesting results were obtained in doped and irradiated samples (Fig. 3 and Fig. 4). After doping silicon samples with palladium atoms, the electrical resistance increases to 322.7 kOhm (1100 ^{0}C) and 137.2 kOhm (1200 ^{0}C). The increase in electrical resistance in doped samples at 1100 ^{0}C is almost 3 times greater than in samples doped at 1200 ^{0}C.

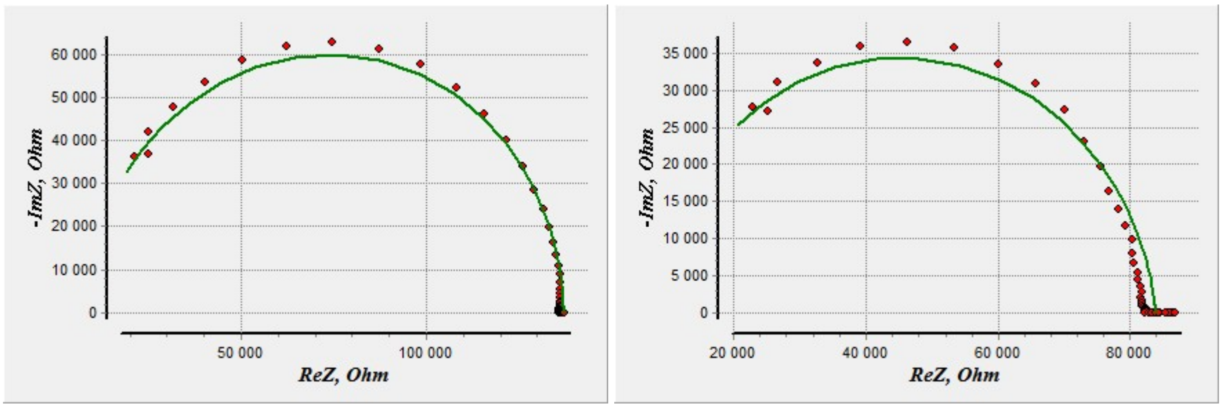


(a) (b)

Figure 3: Hodographs of n-Si samples doped with Pd at 1100 ^{0}C degree: before (a) and after (b) irradiation. The green line is the best fit curve.

In the work (Srivastava et al., 2023) it was shown that electrically active palladium exists in silicon in the form of two independent species. The first, designated Pd_1, is amphoteric and has an acceptor level 0.22 ± 0.01 below the conduction band edge, as well as a donor level 0.33 ± 0.01 eV above the valence band edge. The second species, designated Pd_2, has only an acceptor level 0.32 ± 0.1 eV above the valence band edge. The ratio of Pd1 to Pd_2 embedded in silicon varies from 40 to 5 for diffusion temperatures from 900 to 1200 °C, respectively.

According to (Stoynov & Vladkova, 2005), at doping temperatures above 1150 °C, palladium attains sufficient mobility to diffuse into the silicon crystal lattice and form more stable configurations. Under such conditions, complexes that are energetically close to the equilibrium state are formed, thereby reducing the concentration of deep traps and recombination centers. This process has a beneficial effect on the electrical conductivity of the material, as the number of defects hindering charge carrier transport decreases. Consequently, precise temperature control during palladium doping represents a crucial factor for optimizing the structural and electrophysical properties of the resulting semiconductor.

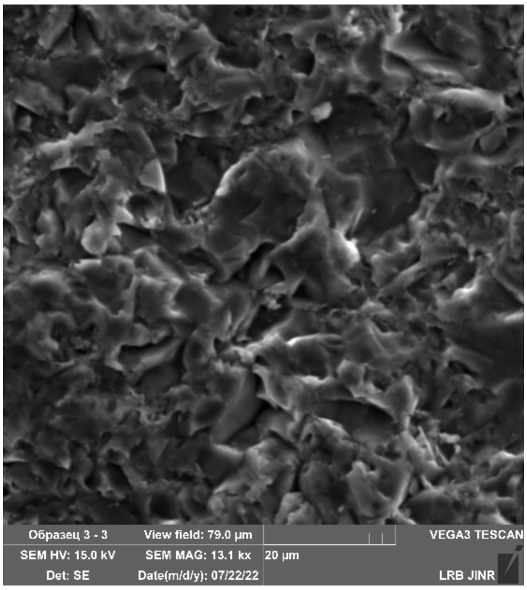


(a) (b)

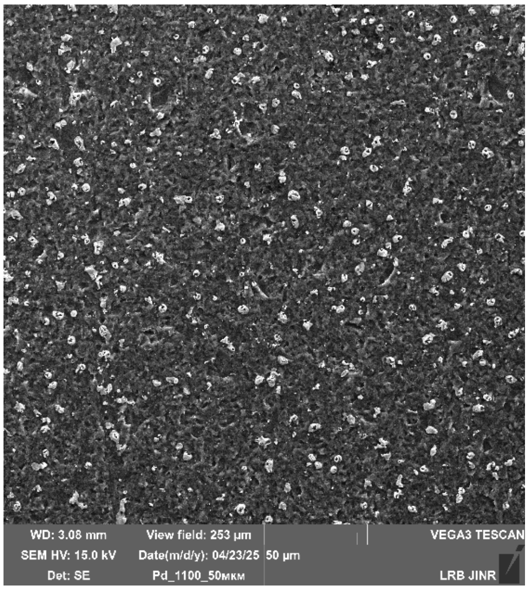
Figure 4: Hodographs of n-Si samples doped with Pd at 1200 ^{0}C degree: before (a) and after (b) irradiation. The green line is the best fit curve.

Palladium introduced into silicon at high temperatures can occupy interstitial or substitutional positions, forming deep levels (traps) and complex defects (Pd–V). The lower the doping temperature, the more unstable and deep defects there are, since palladium does not have time to diffuse into an equilibrium position in the lattice. At 1100 °C, more active traps are formed, which leads to an increase in resistance. At 1200 °C, Pd atoms are distributed more evenly and form less active complexes, and the resistance is correspondingly lower (Utamuradova et al., 2022; Stoynov et al., 2018).

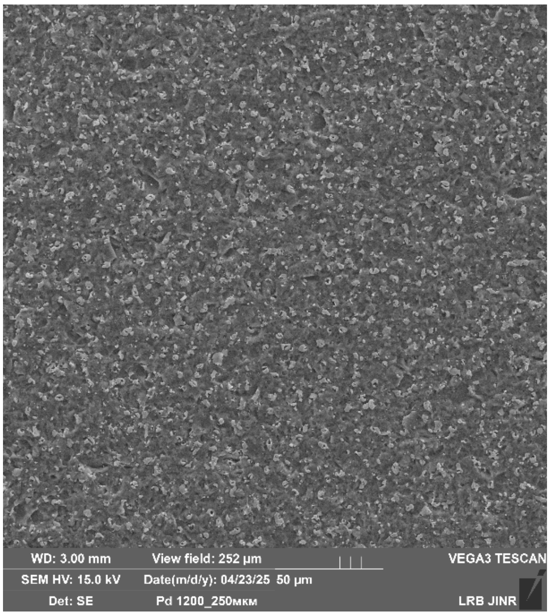
Figure 5 shows scanning electron micrographs of the original and doped samples.



a)



b)

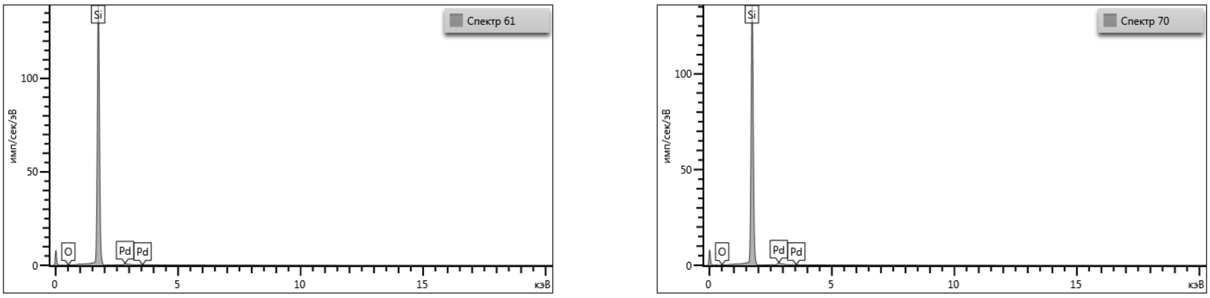


c)

Figure 5: Micrographs of the surface of single-crystal silicon plates before (a) and after doping with palladium (at 1100 °C (b) and at 1200 °C (c))

Micrographs show that doping silicon with palladium atoms at 1100 °C (Fig. 5b) leads to the formation of microstructures with a low concentration, and the introduction of palladium impurity into Si at 1200 °C (Fig. 5c) leads to the creation of smaller microstructures with a high concentration.

Energy dispersive spectra show (Fig. 6) that the number of palladium atoms in silicon doped: at 1100 °C – 3.83 wt.%, at 1200 °C – 5.77 wt.%.



(a) (b)

Figure 6: Energy dispersive spectra of silicon samples doped with palladium: a-1100 °C; b-1200 °C.

Irradiation of the original and doped samples with gamma quanta leads to a change in electrical resistance. The resistance of the original sample increases after irradiation (almost 2 times) and we wrote about this above. But, in the doped samples, we obtained other results (fig. 3b and fig.4b), i.e. irradiation with gamma quanta leads to a decrease in electrical resistance in the doped samples (at 1100 °C - 294.2 kOhm, at 1200 °C - 87.1 kOhm).

In palladium (Pd)-doped silicon samples, γ-irradiation leads to a decrease in electrical resistance, which differs from the behavior observed in pure silicon. This effect may be explained by several mechanisms. First, we assumed, that γ-irradiation may destroy Pd–V (palladium–vacancy) complexes or transform them into electrically neutral states, thereby eliminating their ability to capture charge carriers. In addition, under high irradiation doses, the traps may become filled with electrons, rendering them less active and diminishing their influence on conductivity (Utamuradova & Rakhmanov, 2022; Abiyev et al., 2024).

Second, we expect, that γ-irradiation can alter the energy position of trap levels within the silicon band gap. If these levels are shifted closer to either the conduction or valence band, their carrier capture efficiency decreases. This facilitates the transport of electrons and holes, which in turn reduces the overall resistance of the material. Thus, γ-irradiation in Pd-doped silicon structures can improve the electrical properties by modifying the defect structure (Yunusov et al., 2000).

4 Conclusion

Electrochemical impedance spectroscopy revealed that doping n-Si samples with Pd atoms results in an increase in their electrical resistance (Table 2). Gamma irradiation with an energy of 1.25 MeV at a total dose of 1×10^7 rad caused a nearly twofold increase in the electrical resistance of the original silicon samples. In contrast, for the doped n-Si<Pd> samples, irradiation led to a noticeable decrease in electrical resistance.

Table 2: Limiting values of ohmic resistance for Si samples doped with Pd before and after gamma irradiation

	Resistance [kΩ]		
	n-Si	n-Si<Pd> 1100 °C	n-Si<Pd> 1200 °C
Before irradiation	127.7	322.7	137.2
After irradiation	247.3	294.2	87.1

Changes in the resistance of silicon samples under the influence of doping and γ-irradiation are caused by a complex interaction of the defects that arise. The study showed that the nature and behavior of these defects depend on both the type of external action and the parameters of the technological process.

The main conclusions are as follows:

- In pure n-type silicon, γ-irradiation increases the resistance due to the formation of radiation defects that trap charge carriers.
- Doping silicon with palladium also increases the resistance, especially at low temperatures (1100 °C) of implantation, which is due to the formation of defect complexes.
- In samples doped with Pd, γ-irradiation, on the contrary, reduces the resistance. This may be due to the destruction of existing traps, their transition to a neutral state, or a shift in energy levels, reducing their activity.
- Doping temperature plays a key role in shaping the distribution of palladium defects and their effect on the electrical properties of the material.
- The study of the surface morphology of the doped samples shows that doping silicon with palladium atoms at 1100 °C leads to the formation of a microstructure with a low concentration, and the introduction of palladium impurity into Si at 1200 °C leads to the creation of finer microstructures with a high concentration.

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