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The Effect of Gamma Irradiation on the Thermal Properties of Porous Silicon by Photoacoustic Technique

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This article investigates the impact of gamma irradiation on the thermal properties of porous silicon with varying levels of porosity. Porous silicon is a crucial nanomaterial in modern materials science, widely utilized in electronics, optoelectronics, and various applications. Understanding how its thermal transport properties change under gamma irradiation is of importance for various industries, including military, space, and nuclear technologies, where materials may be exposed to ionizing radiation. For this purpose, we employed the non-destructive photoacoustic method with gas-microphone registration. We assessed thermal conductivity as a function of porosity and irradiation time by simulating the experimental amplitude-frequency dependencies using an appropriate model. Our findings reveal that prolonged gamma irradiation of samples using Iridium-192 with an activity of 50 Curie for up to 20 minutes leads to a decrease in thermal conductivity in porous silicon. This is due to the emergence of defects in the crystalline structure of porous silicon and even its possible amorphization. These defects and alterations in the material's structure restrict the movement of heat carriers, thereby reducing its thermal conductivity. It is worth noting that the most significant change observed in this study is a two-fold reduction in thermal conductivity, particularly evident in samples with the highest level of porosity (60 %). Samples with higher porosity exhibit a stronger response to gamma irradiation because they contain less material within their volume that can conduct heat. The constraints within the crystalline structure of samples with greater porosity create additional barriers to heat transfer, leading to increased vulnerability of the material to radiation and a decrease in its thermal conductivity.

Keywords: Porous silicon, Gamma radiation, Photoacoustic technique, Thermal conductivity.

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1. INTRODUCTION

Research on the impact of ionizing radiation on materials stands as one of the current pressing topics in modern science. Among such materials, porous silicon, particularly, has garnered significant attention due to its unique properties [1]. It has been extensively used as a material for orienting substrates that serve for the growth of various nanostructures in electronics and optoelectronics and as a wide-gap material for complementary metal-oxide-semiconductor (CMOS) technologies, photodetectors and solar cells, one-dimensional photonic crystals, chemical and biological sensors, etc [2-4]. These devices find applications in various industries, including military and space technologies and nuclear power engineering, where they may be exposed to ionizing irradiation [5-7]. Furthermore, analyzing the effects of ionizing irradiation on the structure can lead to the development of methods for controlled tuning of thermal transport within desired limits, a crucial aspect in confined-space thermal engineering [6, 8].

The main goal of this study is to reveal the impact of gamma irradiation on the structure of porous silicon and analyze changes in its physical properties depending on the duration of irradiation. To achieve this goal, we employed the photoacoustic (PA) gas-microphone technique. This method is a reliable and non-destructive tool that has proven its effectiveness in diagnosing porous semiconductor structures [9, 10]. The fundamental principle of the method lies in the registration of acoustic waves generated within the sample due to the absorbed irradiation energy. The amplitude, shape and phase of acoustic signal provide insights into changes in the internal structure of the material and enable the assessment of its thermophysical properties, including thermal conductivity.

In this regard, research into the influence of gamma irradiation on the structure and thermophysical properties of porous silicon using the photoacoustic gas-microphone method holds great significance for both scientific understanding and technological advancements. In the following sections of our article, we will delve into the research methodology, present the results of the analysis of thermophysical changes in porous silicon after irradiation, and investigate potential mechanisms explaining the observed effects.

2. DESCRIPTION OF OBJECTS AND INVESTIGATION METHODS

2.1 Samples Preparation

In this research, we analysed structures composed of a layer of porous and monocrystalline silicon. To create nanoporous silicon (PSi) samples, we employed the elec-

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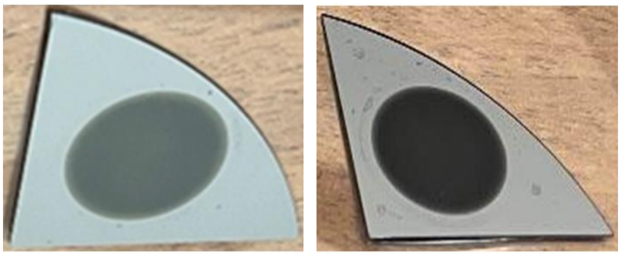
trochemical etching process on boron-doped (boron concentration approximately 10^{19} atoms/cm^3) monocrystalline silicon wafers with <100> crystal orientation. The thickness of these wafers was approximately 500 μm.

As an electrolyte, we used a mixture of concentrated hydrofluoric acid (HF, 49 %) and pure ethanol in a 1:1 ratio. After synthesizing the structures, they were thoroughly cleaned with ethanol and distilled water, and then air-dried at room temperature to remove any remaining electrolyte from the pores. The porosity of the PSi samples and the thickness of the porous layer were controlled by varying the current intensity and etching time. The thickness of the porous layer in the samples was measured both optically and by scanning electron microscopy. Porosity was monitored through gravimetric measurements. As a result, four series of samples were obtained, each containing a porous silicon layer of 3 cm^2 (see Fig. 1) with porosities ranging from 30 % to 60 % in increments of 10 % on the surface of monocrystalline wafers (see Table 1).

Experimental PSi samples were irradiated at room temperature with gamma radiation. The radiation source used was Iridium-192 with an activity of 50 Ci. The absorbed radiation dose was determined by the duration of irradiation. In this work, the irradiation time ranged from 1 to 20 minutes. The visual inspection of the sample surfaces and the measurement of their porous layer thickness through microscopy revealed no discernible alterations as a result of their exposure to radiation.



a b

Fig. 1 – Typical photos of porous silicon samples with 40 % (A) and 60 % (B) porosity, synthesized on a monocrystalline silicon substrate

Table 1 – Description of the special paragraph styles

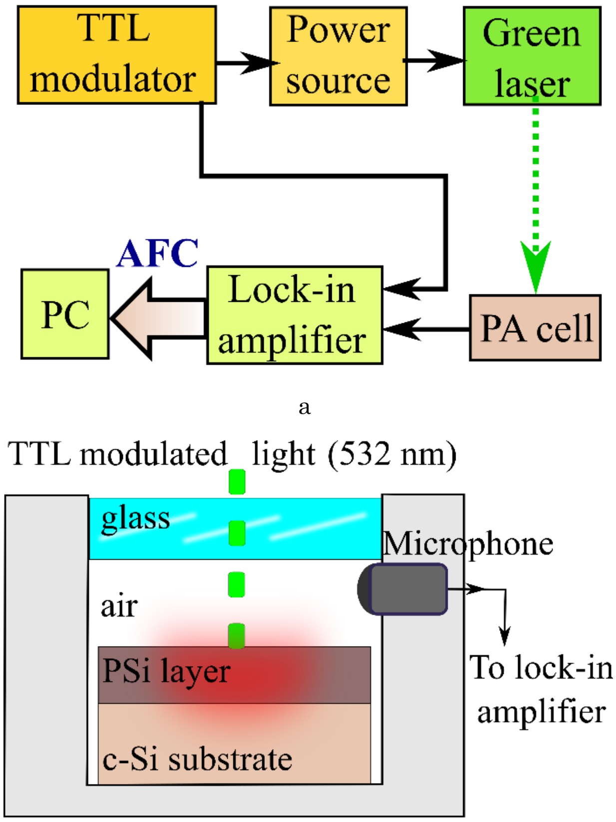
Series of sample	Current density, mA/cm^2	Etching time, min	Porosity, %	PSi thickness, μm
A	10	5	30 %	50
B	40	15	40 %	45
C	70	15	50 %	67
D	150	10	60 %	74

2.2 Experimental Set-up

We conducted thermal conductivity measurements for the PSi samples using a traditional photoacoustic approach involving a gas-microphone setup (see Fig 2 a). To carry out these measurements, we employed a photoacoustic cell equipped with an optically transparent window and a condensed microphone Panasonic WM-61A, in-build inside the PA cell (see Fig. 2 b).

In order to induce a photoacoustic signal, we used a green laser emitting at 532 nm. The modulation of the laser was achieved through adjustments in its power source, generating square wave modulation. The initial power of the laser irradiation was approximately 80 mW, and the diameter of the output beam focused on the surface was 1 mm. The PA signal for each series of samples were experimentally recorded within the 10-2500 Hz by lock-in nanovoltmeter Unipan 232B.

Before conducting experiments, we calibrated the photoacoustic cell using reference samples with well-established thermal and optical properties to ensure measurement accuracy and mitigate unwanted signals from factors like cell properties (heat-sink effects, optical properties), microphone characteristics (sensitivity losses, design features), and electrical circuits in our equipment. Additionally, acoustic resonances inherent to the photoacoustic cell, influenced by its dimensions, and other sources like the microphone diaphragm, were considered. We also addressed signal-to-noise ratio changes at higher modulation frequencies. Our careful analysis and calibration, especially at low and high frequencies, ensured alignment with the theoretical model, providing accurate data.



b

Fig. 2 – Schematic representation of the experimental photoacoustic setup (a), photoacoustic gas-microphonic cell in a classical configuration (b)

3. RESULTS AND DISCUSSION

As a result, for each series of samples, we obtained informative Amplitude-Frequency Dependences (AFD) of PA signal. For instance, a comparison of AFD for PSi samples with different porosity before and after the exposure to gamma radiation for 20 minutes is provided in Fig. 3 a and Fig. 3 b, respectively.

To analyze the data, we employed the framework of thermal wave formalism. Within this theoretical framework, the thermal perturbation generated by the modulated light can be expressed as a rapidly decaying thermal wave. This wave can be characterized by its wavelength, which plays a crucial role in understanding the material's thermal properties.

$$\lambda_{th} = \sqrt{D_T / \pi \cdot f} \tag{2.1}$$

where λ_{th} is wavelength of thermal wave, D_T is the thermal diffusivity of the sample.

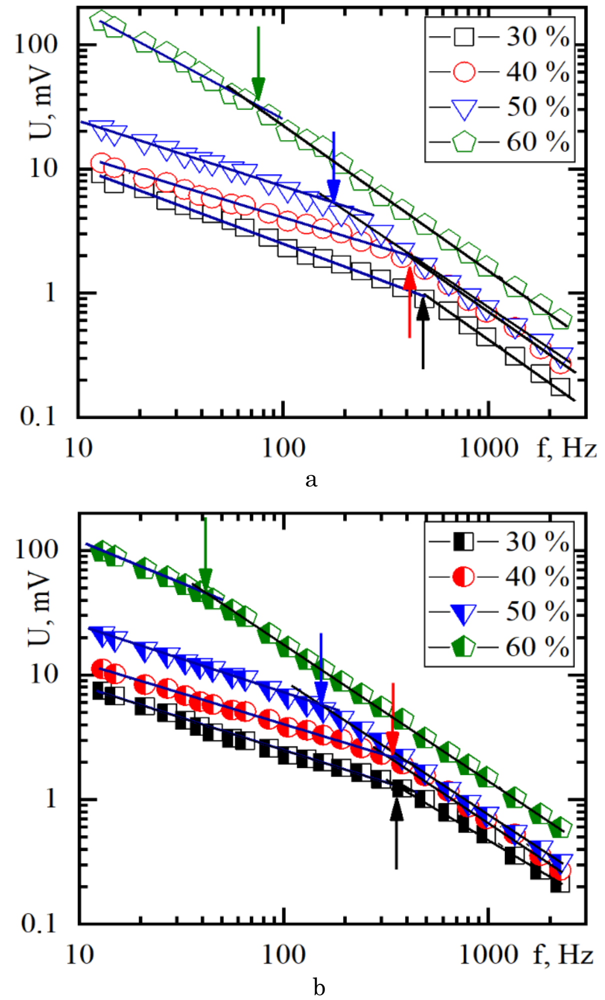


Fig. 3 – Experimental AFC of the photoacoustic signal for the initial PSi samples with various levels of porosity on a monocrystalline silicon substrate (a), AFC of the PSi samples subjected to 15 minutes of gamma irradiation with Iridium-192 (b). The arrows indicate the positions of the "critical frequencies" on the AFCs of the samples

When considering higher frequencies (where the thermal diffusion length, λ_{th}, is smaller than the thickness of the porous silicon layer, l_{PSi}), the thermal perturbation is primarily confined to the top layer. However, as the modulation frequency decreases, the wavelength becomes larger than l_{PSi}, and the thermal behavior starts to be influenced by the thermal properties of the underlying silicon layer.

Consequently, in the amplitude-frequency dependencies of the photoacoustic signal plotted on a double logarithmic scale, a bend occurs at a frequency corresponding to $\lambda_{th} \approx l_{PSi}$. This bending frequency, often referred to as the "critical frequency" in the literature, provides a means to estimate the thermal diffusivity and thermal conductivity of porous silicon using the following expressions:

$$D_T = \pi \cdot f_c \cdot l_{PSi}^2 \tag{2.2}$$

where f_c is a critical frequency.

The analysis of the AFC for all samples revealed a clear dependency of the critical frequency on porosity and the irradiation time (see Table 2). Therefore, according to formula (2.2), it is possible to determine the thermal diffusivity of the samples.

Table 2 – Critical frequency values for the PSi samples with different porosity and gamma irradiation time

Samples No	Porosity, %	Irradiation time, min	Critical frequency, Hz
A0	30 %	0	480 ± 11
A1		1	435 ± 18
A2		5	425 ± 25
A3		10	365 ± 20
A5		20	365 ± 25
B0	40 %	0	413 ± 20
B1		1	375 ± 20
B2		5	365 ± 21
B3		10	379 ± 05
B4		15	350 ± 10
B5		20	350 ± 10
C0	50 %	0	176 ± 13
C1		1	180 ± 15
C2		5	185 ± 30
C3		10	135 ± 03
C4		15	160 ± 12
C5		20	150 ± 10
D0	60 %	0	76 ± 20
D1		1	75 ± 10
D2		5	55 ± 03
D3		10	42 ± 20
D4		15	49 ± 05
D5		20	40 ± 03

Furthermore, we can estimate their thermal conductivity values by recalculating thermal diffusivity and making a simplified assumption of constant volumetric heat capacity:

$$\chi = D_T \cdot C \cdot \rho_{PSi} \tag{2.3}$$

where C is a specific heat of PSi, $\rho_{PSi} = \rho_{Si}(1-\varepsilon)$ is a density of PSi layer, ε is porosity of the PSi layer.

This approach offers a rough approximation to provide a general understanding of the order of magnitude. The corresponding values of thermal conductivity of the samples as functions of porosity and irradiation time are depicted in Figure 4.

First, we observe a significant decrease in the thermal conductivity of the samples with an increase in their porosity. The low thermal conductivity of PSi can be attributed to a combination of factors. The porosity (free volume) of PSi plays a substantial role. The presence of pores within the material limits the efficient transmission of heat. Furthermore, the formation of nanoscale dendrites within PSi introduces phonon scattering, which further reduces thermal conductivity. Additionally, the heat flow in PSi encounters challenges due to percolation effects. Heat must traverse through narrow constrictions within the dendritic structures, causing a reduction in thermal conductivity. This reduction is especially pronounced as porosity increases, and it has been shown that thermal conductivity crucially decreases with porosity [11].

Secondly, from Figure 4, it can be observed that an increase in the irradiation duration of PSi samples leads to a reduction in their thermal conductivity. This phenomenon can be explained by several factors.

Primarily, gamma radiation is believed to influence the structure of the porous material, potentially leading to the formation of defects and possible amorphization of porous silicon, similar to the findings of the ion irradiation impact study described in references [6, 8, 12, 13]. These defects can restrict the movement of heat carriers, thereby affecting the material's thermal conductivity. Similar results have been observed in previous studies, when irradiating PSi with other irradiation sources. Additionally, changes in thermal transport can be influenced by the effect of samples oxidation. This behavior can be attributed to the fact that, in the initial stages the oxidation causes a decrease in the size of the silicon crystallites, consequently resulting in a reduction of thermal conductivity.

It should be noted, that the most significant change in thermal conductivity (a two-fold reduction) is observed in samples with the highest porosity (60%). This can be attributed to the increased interaction of ionizing radiation with the nanostructured material under conditions of its pronounced confinement due to the absence of a significant amount of material.

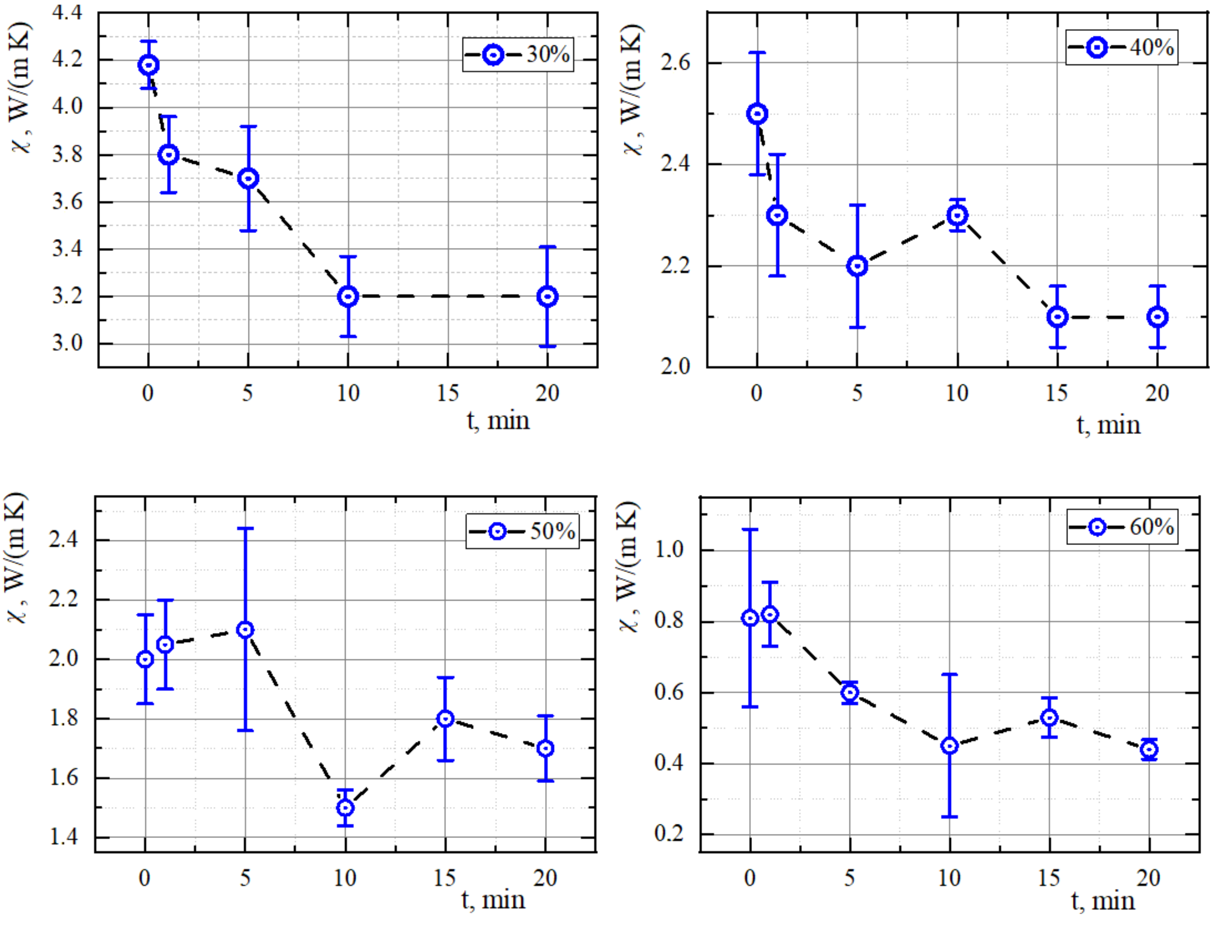


Fig. 4 – Graphs depicting the relationship between the thermal conductivity coefficient and irradiation time for samples containing a layer of porous silicon with porosities of 30 %, 40 %, 50 %, and 60 %, respectively

4. CONCLUSIONS

In this study, we investigated the thermal transport properties of gamma-irradiated PSi nanostructures. Employing the photoacoustic approach, revealed a significant reduction in thermal transport properties within the irradiated structures, indicating a potential for modifying the thermophysical parameters of PSi nanostructures, broadening their application prospects.

Notably, the most substantial change in thermal conductivity, a two-fold reduction, was observed in samples with the highest porosity (60 %). This alteration results from heightened ionizing radiation interaction with the nanostructured material, particularly in conditions of pronounced confinement due to the scarcity of material.

In conclusion, this study uncovers the intricate interplay between porosity, gamma irradiation, and the thermal properties of porous silicon, providing crucial insights for further investigations into alterations in thermal transport stability of the studied nanostructures and potential applications of gamma radiation in modifying the thermophysical characteristics of nanostructured materials.

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Дослідження впливу гамма-опромінення на теплові властивості поруватого кремнію за допомогою фотоакустичного методу

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В статті досліджується вплив гамма-опромінення на теплові властивості поруватого кремнію з різним рівнем поруватості. Цей матеріал є важливим об'єктом в сучасному матеріалознавстві та знаходить широке застосування в електроніці, оптоелектроніці та різних областях. Розуміння того, як змінюються його теплові властивості під впливом гамма-опромінення, є важливим для різних галузей промисловості, включаючи військову, космічну та ядерну технології, де ці матеріали можуть бути піддані впливу іонізуючого випромінювання. Для реалізації нашого дослідження ми використовували неруйнівний фотоакустичний газо-мікрофонний метод. В результаті було оцінено теплопровідність зразків в залежності від їх поруватості та тривалості опромінення на основі аналізу експериментально отриманих амплітудно-частотних залежностей фотоакустичного сигналу відповідною моделлю. Наші результати показують, що гамма-опромінення зразків ізотопом іридію-192 із активністю 50 кюрі протягом до 20 хвилин, призводить до зниження теплопровідності в поруватому кремнії. Це пов'язано з появою дефектів у кристалічній структурі пористого кремнію та навіть його можливою аморфізацією опроміненням. Ці зміни в структурі матеріалу обмежують рух теплоносіїв, що в свою чергу призводить до зменшення його теплопровідності. Важливо відзначити, що найсуттєвіша зміна теплопровідності (до 2 разів), спостерігається у зразках із найвищим рівнем поруватості (60%). Зразки з вищою поруватістю демонструють більш виражену відповідь на гамма-опромінення через менший обсяг матеріалу, який може передавати тепло. Ймовірно, це пов'язано з тим, що при однакових дозах опромінення відсутність більшої кількості матеріалу у зразків з більшою поруватістю створює додаткові перешкоди для передачі енергії, що призводить до більшої вразливості матеріалу до радіації.

Ключові слова: Поруватий кремній, Гамма випромінювання, Фотоакустичний метод, Коефіцієнт теплопровідності.