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Optimal ranges determination of morphological parameters of nanopatterned semiconductors quality for solar cells

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ABSTRACT

Purpose: The paper aims to determine the values of the main morphological characteristics of nanopatterns, which can be considered as the reference for use as surfaces of solar cells.

Design/methodology/approach: The article uses an approach based on the definition of reference indicators of nanopatterns for solar cells by analysing the main parameters of solar cells and comparing them with the possible values of morphological parameters. Correlations of pore radius and visible wavelength, porosity and visible range, wavelength of de Broglie, nanopatterned layer thickness and charge carriers diffusion length, etc., are analysed. Compliance verification of morphological characteristics of nanopatterns with the specified criteria was performed on the example of porous silicon layers.

Findings: The conducted research allowed to define the basic values of morphological parameters of porous nanopatterns, namely porousness, pore size (effective diameter), the thickness of the porous layer, and form factor. Reference ranges of morphological parameters of nanopatterns formed on the surface of semiconductors for applications in solar cells are established.

Research limitations/implications: The article is devoted to the choice of optimal morphological characteristics of porous nanopatterns on the surface of semiconductors for solar cells. However, for solar cells, other types of nanopatterns can also be applied, for which it is also necessary to develop methods for selecting optimal parameters. Moreover, the prospect of research on this topic is to check the intrusion into a certain range of values of real nanopatterns formed on the surface of semiconductors.

Practical implications: In the article the methodology allowing to choose optimal values of morphological parameters of nanopatterns for its application for solar cells is considered. Such studies are of great practical importance for the production of high-quality solar cells based on nanopatterned semiconductors.

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Originality/value: The article for the first time considers the choice of the nanopattern type and the ranges of morphological parameters in terms of quality assurance of the final product – the solar cell. It is determined that it is necessary to take into account such factors as porousness, pore size, thickness of the porous layer and roundness. A range of optimal values is selected for each of the indicators.

Keywords: Nanopatterns, Solar cells, Morphological parameters, Quality

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PROPERTIES

1. Introduction

The most promising method of alternative energy is the solar photovoltaic conversion due to the existing advantages [1-3]:

- direct conversion of light quantum energy into electrical energy;
- the variety of the elementary base for the manufacture of solar cells;
- ability to create modular systems of different capacities;
- the possibility of using concentrated solar radiation;
- noiselessness;
- flexibility in application;
- sustainability.

Among the disadvantages of using solar energy, the following are most often allocated [4,5]:

- high cost value of solar panels;
- electricity generation during daylight hours only;
- dependence on climatic conditions;
- the need for large areas for the construction of photovoltaic power station;
- problems with energy accumulation storage;
- imperfection of technology and low efficiency factor, etc.

It is the imperfection of the technology for creating photo-emissive converters (PECs) and their low efficiency factor that is the main deterrent to the global replacement of traditional energy with renewable energy [6,7]. That is, there is a need to develop innovative technologies improving the efficiency factor and other electrical characteristics of solar panels.

In this regard, relevant are the studies devoted to finding ways to improve the technological processes of manufacturing photo-emissive converters [8,9]. When developing appropriate technological solutions, it is necessary to take into account the characteristics of materials used as raw materials for PECs.

Solid-state single-crystalline Si-Solar cells with p-n transitions are the basis of the most widespread commercial photovoltaic devices [10,11]. The well-developed technology of obtaining and processing single-crystalline silicon allows us to expect the retention of key positions for solar cells based on it in the near future [12].

Silicon is the second most widespread element in the Earth's crust (35%) after oxygen. It is the main material for photovoltaic conversion of solar radiation spectrum in the range from ultraviolet to near reared [13], but it can absorb a small part of solar radiation. Through this, the efficiency for ideal silicon solar cells can reach about 30% (at 300 K) [14].

Due to the high refraction angle index of silicon ($n = 3.5$), a significant part of solar radiation is reflected from the surface of the photovoltaic converter (the reflection value can be ~35%) and, therefore, does not contribute to the generation of electron-hole pairs [15]. This leads to a decrease in the efficiency factor of such converters [16]. As a rule, the problem of reflection reduction is solved by applying anti-reflective coatings to the surface of solar cells [17]. The use of such coatings leads to an increase in the conversion efficiency factor, an extension of the service life and an improvement in the electrophysical and operational characteristics of photovoltaic converters [18]. The high refraction angle index of crystalline silicon in the (300-1100) nm creates large optical losses, which can be reduced by an antireflection coating [19]. Although highly efficient double and triple anti-reflective coatings are available, most industrial crystalline silicon solar cells use a simple and inexpensive single-layer ARC with relatively poor anti-reflective properties [20].

Among the main directions of overcoming the problem of imperfection of photo-emissive converters can be identified as follows:

- improvement of technological process of elementary base preparation [21];
- use of anti-reflective coatings for PECs [22];
- search for new materials for solar panels [23].

Efforts in the search for new materials for PECs are directed towards the use of nanopatterned semiconductors based on crystalline [24,25].

The use of porous silicon for solar panels was investigated by the authors of the work [26]. It is proved that an increase in conversion efficiency (up to 20%) is achieved for por-Si solar cells compared to cells without a porous layer.

Electrochemical etching methods are traditionally used to form a nanopatterned layer on the surface of semiconductors [27,28]. Electrochemical etching leads, first of all, to the change in the morphology of the surface layers of the semiconductor [29,30]. The change in morphology occurs due to etching of the upper layers and formation of nanorelief on the surface [31,32]. Dense pore channels or pyramids with different plain inclinations are formed on the surface of semiconductors [33,34].

However, despite the progress made in the nano-patterning of semiconductors, the significance of the characteristics of nanopatterns that need to be provided for their application in solar energy has not yet been determined.

The aim of the paper is to determine the values of the main morphological characteristics of nanopatterns, which can be considered as reference for use as surfaces of solar cells. This will allow to control the synthesis process of the nanopatterns to form them with predetermined properties.

2. Methodology and results of the study

The use of nanopatterns as materials for the PECs provides the introduction of additional steps in the standard technology for the creation of photo-emissive converters.

Hard semiconductor solar panels are based on silicon, gallium arsenide and indium phosphide. Each of these semiconductors has its pros and cons for the use in PECs (Tab. 1).

Production of solar panels based on nanopatterned semiconductors consists of three main blocks:

- nanopatterning of semiconductors;
- production of solar panels based on nanopatterned semiconductors;
- production of solar panels – mounting them from photovoltaic cells.

Table 1.
Parameters of semiconductors used to create a PEC based on them

Characteristics	Si	GaAs	InP
Gap energy Eg, eB	1.11	1.42	1.38
Charge number, Z	14	32	32
Radio resistance, dose g-emission, Gr	$(10^2\text{-}10^3)$	$(10^5\text{-}10^6)$	$(10^6\text{-}10^7)$
Maximum of operating temperature of PEC, T, °C	70	150	200

The principal in all three stages of the process is to control the products quality at each stage. Quality control should include technological process control, performance results control, and output control of valuable parts. This is ensured by testing samples and sorting out the defective ones.

In the current study, the methodology is represented allowing to choose optimal values of morphological parameters of nanopatterns for its application for solar cells.

Compliance verification control of morphological characteristics of nanopatterns with the specified criteria was performed on the example of porous silicon layers. The main morphological characteristics of porous silicon samples and ways to ensure compliance with the established quality criteria are considered. The main morphological characteristics include the average pore diameter, the porousness, the thickness of the porous layer and the pore form factor. The procedure for evaluating the quality of the samples presented below can be extended to other nanopatterns synthesized on the surface of semiconductors using electrochemical etching technology.

The reference values of the quality criterion of the nanopatterned semiconductor surface are determined on the basis of the functional purpose of the material. With an overview on this, it is appropriate to select the pore size so that the pore radius is not less than the wavelength of the visible range, that is, the condition must meet the requirement:

$$\frac{dp}{2} \geq \lambda,\ \lambda \subseteq [380,\ 790] \tag{1}$$

where d_p – pore diameter, nm; λ – length of visible light range, nm.

On the other hand, to extend the range of light absorption becomes possible by using patterns in which the effects of size quantization are manifested, i.e. the changes of the thermodynamic, kinetic, optical and other properties of the material when at least one of its geometric sizes is proportional to the de Broglie wavelength of electrons. The quantum dimensional effect is related to the quantization of the charge carriers energy the movement of which is limited to one, two or three directions. The de Broglie wavelength is determined from the ratio [35,36]:

$$\lambda_b = \frac{h}{p} \tag{2}$$

where p – photon momentum.

Since we have chosen massive pores, they cannot serve as sources of quantum-dimensional effects. These sources will serve as interporous separating partitions – nano-crystallites separating the pores. For the quantum-dimensional effect to occur, the cross-section between the pore partitions (h_p) must be of micrometre scale. That is to meet the requirements:

$$hp \subseteq [2,\ 15]\ \text{nm}. \tag{3}$$

The determination of the values of the variable h_p in a given range is difficult because of the very small values. Therefore, the calculation of the average cross-section of the interporous space is advantageously carried out by mediate methods, for example, the density of pores or porousness. The denser the pores are pressed against each other (large porosity), the less important is the cross-section of the interporous space. So the porousness must be within:

$$P \subseteq [75,\ 90]\%. \tag{4}$$

The value $P = 90\%$ is critical, because when this value is exceeded, the porous layer becomes very brittle and crumbles.

In order to ensure the permeability of the light beam in the pore it is necessary to form cylindrical mutual parallel channels of pores, which are oriented perpendicular to the surface of the crystal. Cylindrical pores have a circle in its cross section, so the form factor must meet the requirements:

$$Fp \to 1. \tag{5}$$

The thickness of the porous layer (pore depth l) should be determined from the following considerations. The pore depth should be such so that the distance from the pore bottom to the p+-layer (L) is approximately equal to the diffusion length of the charge carriers:

$$L \,\square\, Ld\,. \tag{6}$$

The diffusion length is a physical parameter described by the formula [37, 38]:

$$Ld = \sqrt{Dt}\,, \tag{7}$$

where D is the diffusion coefficient of the charge carrier in a particular semiconductor, t is its life span.

The diffusion length is approximately 1 mcm and is a measurable unit [39,40]. Therefore, the optimal pore depth will be determined by the thickness of the n-layer [41,42].

Thus, it is possible to allocate the reference values of the porous pattern of the semiconductor for the PEC (Tab. 2).

Table 2.
Reference values of the porous layer on the surface of semi-conductors, which can improve the efficiency of the PEC

Index	Measurement Unit	Reference value, range
Pore diameter	nm	200-400
Porousness	%	75-90
Form factor		1
Thickness of the porous layer	mcm	20-35

To describe one of the main characteristics of the pore form, the ImageJ program uses the concept of the form of circularity factor, which is calculated using the formula:

$$Fp = \frac{4\pi S}{p^2} \tag{8}$$

where S is the pore space and p is the pore perimeter.

The value of the form factor $Fp = 1$ indicates that the pore section is an ideal circle [43]. The closer the circularity value is to 0, the more elongated or deformed the pore section will be.

3. Experiment and research results

Sets of n-type crystalline silicon plates were used for the experiment. The crystals were grown using the Czochralski process, being cut into 1 mm thick plates and polished on both sides. Porous surfaces were formed by electrochemical etching in an acid solution. The experimental conditions are shown in Table 3. Using different processing modes, it is possible to control the parameters of the resulting structures. The purpose of the experiment is to establish silicon processing modes, in which a porous layer with reference values is formed on the surface.

Table 3.
Experiment conditions

Sample number	The etching time, t, min.	The current density, j, mA/cm^2	Electrolyte
1	5	75	*HF: H_2O=1:1*
2	10	75	*HF: H_2O=1:1*
3	15	75	*HF: H_2O=1:1*
4	20	75	*HF: H_2O=1:1*
5	25	75	*HF: H_2O=1:1*
6	5	100	*HF: H_2O=1:1*
7	10	100	*HF: H_2O=1:1*
8	15	100	*HF: H_2O=1:1*
9	20	100	*HF: H_2O=1:1*
10	25	100	*HF: H_2O=1:1*
11	5	125	*HF: H_2O=1:1*
12	10	125	*HF: H_2O=1:1*
13	15	125	*HF: H_2O=1:1*
14	20	125	*HF: H_2O=1:1*
15	25	125	*HF: H_2O=1:1*
16	5	150	*HF: H_2O=1:1*
17	10	150	*HF: H_2O=1:1*
18	15	150	*HF: H_2O=1:1*
19	20	150	*HF: H_2O=1:1*
20	25	150	*HF: H_2O=1:1*

The surface morphology was studied using a raster electron microscope. ImageJ and Origin software was used to analyse the morphological properties of synthesized nanopatterns.

Based on the results of raster electron microscopy, it was found that the active pore formation begins if the etching duration is not less than 10 minutes. For the selected type of crystals, the pores formation with a round transverse diameter is observed. Almost all of the samples obtained show a uniform distribution of pores on the surface.

Morphological analysis was performed on the example of sample No. 18 formed in a solution of hydrofluoric acid for 15 minutes. Figure 1 presents the scientific imaging of por-Si obtained using the JEOL-6490 raster electron microscope. According to the visual analysis of the image, the pores have a round shape, most of the pores are isolated from each other, located evenly on the surface.

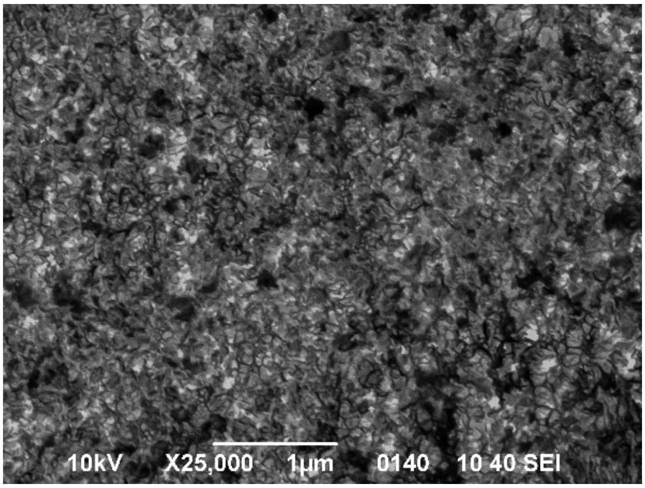

Fig. 1. Morphology of por-Si: HF electrolyte: H_2O=1:1; etching time t = 15 min, current density j = 150 mA/cm^2

The pore distribution by diameter is analysed using the ImageJ software. Figure 2 shows the pore distribution on the surface of sample No. 18 in diameter.

From Figure 2 we see that the values of pore diameters are in a fairly narrow range (Tab. 4).

Table 4.
Parameters of nanopatterned semiconductor layers and its properties

Characteristics being analysed	Physical quantity unit determination	Range of valid values	Relative error (P = 0.95), %
D10	nm	65-100	±7
D50	nm	200-350	±5
D90	nm	80-440	±7

Similar studies concerning the pore size and pore distribution evaluation by diameter value were performed for all samples.

The generalized analysis results of these characteristics for the cross-section of the porous surface of 20 studied samples being synthesized under similar conditions also show an almost complete matching of these characteristics (Tab. 5).

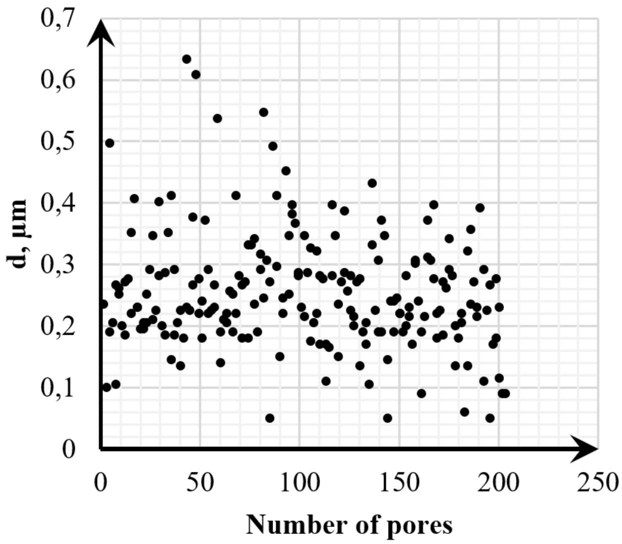

Fig. 2. Histogram of pore spread across the diameter of sample No. 18

Table 5.
Average values for 20 por-InP samples

Sample number	Pore diameter, nm	Porousness, %	Form factor	Thickness of the porous layer, mcm
1	15	3	0.1	0.690
2	37	10	0.9	0.708
3	89	13	1.5	0.793
4	112	29	1.7	0.848
5	115	54	1.7	0.868
6	27	12	5.0	0.767
7	78	22	17.4	0.501
8	85	43	19.2	0.850
9	119	48	21.3	0.558
10	197	17	22.1	0.745
11	89	38	9	0.724
12	187	46	12	0.683
13	241	74	18	0.872
14	312	78	21	0.739
15	338	65	29	0.802
16	398	23	27	0.841
17	275	76	31	0.941
18	397	89	36	0.627
19	429	89	31	0.653
20	441	61	15	0.788

Table 6.
Parameters of nanopatterned semiconductor layers and its properties

Parameter	What effect does it cause?	Advantage
Porousness	Absorption of the major part of light by increasing the effective area	Significant increase in efficiency factor compared to single-crystal and polycrystalline compatibles
	Increase of the band gap width due to the effect of quantum charge retention in microcrystallites	Allows the use of porous layers as a wide-banded illumination-sensitive layer
Pore size	The target range of electromagnetic interference is extended through the shift of the FL peaks to the visible part of the spectrum	The possibility to absorb a wide range of light appears
	Minimization of the reflectivity by light trapping in the pores	Application as anti-reflective coating
Porousness + pore size	Reduced light reflection due to increased surface roughness	Application as anti-reflective coating
Oxidation of surface layers	Chemical and physical inactivity due to the formation of a passivating oxide layer	Reduced sensitivity to surface contamination, loss reduction

The average pore diameter in the interval range is 6 samples (from 13 to 18). Among these samples, the number of samples within 75-90% have samples of 5 samples (13, 14, 15, 17, 18). Among these 5 samples, the thickness of the porous layer is within the established limits in samples 14, 15, 17. Two times the samples have a form factor meeting the established reference values (No. 15 and 17). That is, these samples can be considered as high-quality, its morphological parameters fully correspond to the standard ranges of the porous surface values, which can be used in solar cells. Samples No. 13, 14, 18 can be considered conditionally as high-quality.

The analysis, on the one hand, shows the method of selecting quality samples, on the other hand, allows you to set the technological modes in which quality samples are formed, the indicators of which correspond to the standard ranges of values. Thus, according to the results in Table 5, it can be concluded that high-porosity silicon with massive pores (200-400 nm) must be formed at a current density (125-150) mA/cm^2 for 10-25 minutes.

The formation of porous layers on the surface of semiconductors leads, first of all, to an increase in the effective area by thousands and tens of thousands of times (depending on the degree of porousness). Obviously, this fact leads to an increase in the efficiency factor (from 20% and above) of solar modules with the use of nanopatterns (Tab. 6). Due to the shifts of the photoluminescence peak, the target range of electromagnetic interference is extended. The porous profile of the surface provides a significant increase in roughness allowing the use of porous layers as an anti-reflection coating.

The above (Tab. 6) advantages of using nanopatterned semiconductors over single-crystal semiconductors make them indisputable candidates as the main material for PEC.

4. Conclusions

1. Thus, the main values of the morphological characteristics of porous nanopatterns, which can serve as reference values for the use of these nanopatterns as a material of photo-emissive converters, are determined.
2. The porousness, pore size (effective diameter), the thickness of the porous layer and the form factor (roundness of the pores) are related to the major morphological characteristics.
3. It is established that the optimal value can be considered the porous layers with an average pore diameter in the range 200-400 nm, surface porousness within 75-90% and the thickness of the porous layer from 20 to 35 nm.
4. In cross section, the pores should have a circle.
5. Compliance verification control of morphological characteristics of nanopatterns with the specified criteria was performed on the example of porous silicon layers. The main morphological characteristics of porous silicon samples and ways to ensure compliance with the established quality criteria are considered.

Providing such indicators of the morphology of porous layers will let the use of an effective coating of photo-emissive converters.

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