Показана перспективність створення ефективних сонячних елементів з використанням багатофункціональних мультитекстур пористого кремнію (ПК). Теоретично досліджено зв'язок між діаметром пори dP, пористістю P та областю питомої поверхні S. Багатофункціональні мультитекстури були створені на фронтальній поверхні фотоелектричних перетворювачів. Параметри ФЕП були підтверджені вольт-амперними характеристиками СЕ та вимірами ефективності отриманих СЕ

Ключові слова: сонячний елемент, пористий кремній, фотоелектричний перетворювач, ефективність перетворення, багатофункціональна мультитекстура

Показана перспективность создания эффективных солнечных элементов с использованием многофункциональных мультитекстур пористого кремния (ПК). Теоретически исследована связь между диаметром поры dP, пористостью P и областью удельной поверхности S. Многофункциональные мультитекстуры были созданы на фронтальной поверхности фотоэлектрических преобразователей. Параметры ФЭП были подтверждены вольт-амперными характеристиками СЭ и измерениями эффективности полученных СЭ

Ключевые слова: солнечный элемент, пористый кремний, фотоэлектрический преобразователь, эффективность преобразования, многофункциональная мультитекстура

UDC 621.315.592
DOI: 10.15587/1729-4061.2017.96906

DEVELOPMENT OF EFFICIENT SOLAR CELLS WITH THE USE OF MULTIFUNCTIONAL MULTITEXTURES

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1. Introduction

The widespread introduction of solar cells (SC) with different levels of capacity, as a power source for electronic devices, is the most promising way to use solar energy. This is caused by the depletion of energy raw materials on Earth and continuous growth of the need of humanity in cheap electrical energy.

To date, more than 2 billion people on the planet still depend on gas, wood, coal and oil for cooking and heating the premises. This leads to significant negative consequences for the health of people, to their environment, economic development, and sometimes wars between states. And in the next decades the energy producers will face a shortage of natural fuels (oil, gas, coal), as well as such problems as catastrophic environmental pollution, which is predetermined by burning these fuels, and potential danger of nuclear power. Therefore, there is a necessity to obtain cheap energy by using renewable power sources with minimal impact on the environment.

This became the impetus for the development of high-power solar energy that could compete with traditional methods for the generation of electricity to satisfy energy needs of humanity. Today, photoenergy is one of the most promising sectors of modern industry, which rapidly develops, and in which one of the largest increases in the production of electricity has been obseved in recent years. A whole series of advantages, specific to photoenergy, defines the areas of research, while demand and problems associated with traditional energy sources determines the scope of government programs that stimulate production. The need to reduce the cost, to improve the photovoltaic parameters of SC leads to the development and improvement of technologies for creating new structures of SC. Silicon is the most common semiconductor material in nature, and the most promising for use in photoenergy. Therefore, the most interesting are the studies in the field of creating the silicon SC, thus the development of specialized silicon technological equipment for their production is relevant now. Over the last decade, most ground-based photovoltaic systems in industry (provided the system operates for 20–30 years or longer) are made based on crystalline and multicrystalline silicon with the average value of conversion efficiency at 16 % –17 % – 19 %.

That is why the so-called "alternative" or "non-traditional" energy engineering is gaining traction now, which uses virtually inexhaustible resources [1]. On the European continent, from 2009 to 2016, the cost of photovoltaic electricity dramatically decreased from 50 to 20 eurocents and in some regions to 7 eurocents. In the Arab countries, reduction in the cost of photovoltaic electricity led to even lower price of 5 eurocents. All this was registered in the latest report of the EU for 2016.

Photoenergy is the most elegant means to generate electricity without movable parts, gas release or noise. And this is all possible when converting an infinite amount of solar energy. It became the impetus for the development of high power solar energy that could compete with traditional methods in the generation of electricity to meet the energy needs of humanity.

In order to reduce the cost of manufacturing photovoltaic systems of high power, different approaches are often used. One of them is to improve the efficiency of SC and reduce the cost for the fabrication of SC. In line with such approaches, the basis of high-power solar energy was formed by silicon SC with diffusion p/n-transition, which achieved the highest value of efficiency. However, despite their high efficiency, the cost of these SC remains high as a result of significant complexity of their technological implementation.

The introduction of silicon functional porous materials to one of the most promising directions of SC structures will make it possible to control their conversion efficiency. At the same time, it is possible to reduce the cost of constant of 1 kW of power. In this case, the efficiency of conversion of SC consequently increases under condition of reducing their cost. Functional porous materials can be received in the form of coatings with a sufficiently large range of porosity, with a diameter of pores from nanometers to tens of micrometers. Underlying their obtaining is the hybrid technology, which is based on both electrochemical and the chemical etching.

Important is the clean conversion of sunlight into electricity as well as their heat. The introduction of non-traditional energy sources becomes more profitable, both from an economic and environmental point of view. There is an increased interest to both theoretical and practical developments in the field of photoelectric converters of solar radiation.

All SC have different structure. The only drawback is the high price of such structures, at high degradation. The creation of multi-functional multitexture on the front surface of a photoelectric converter using hybrid technology of obtaining the PSwill allow reducing the cost and improving the technical and photovoltaic parameters of SC.

2. Literature review and problem statement

Most of the textured surfaces demonstrate satisfactory anti-reflective coatings only in a limited range of wavelengths (500–900 nm), whereas in the near infrared and ultraviolet range the reflectivity may be decreased only by 60–90 %. Microtextured surface of the silicon, formed under the action of femtosecond pulsed laser, has a high indicator of absorption in a wide range of wavelengths (250–2500 nm), which allows reducing the reflectivity below 10 % [2]. However, the above technologies employ sophisticated equipment that predetermines high cost of the resulting product. It is possible to reduce the cost of SC along the way of forming the layers of porous silicon (PS) of the "Black Si" type by using electrochemical etching [3, 4]. All these factors need to be considered when choosing the technological method for obtaining the front surface of SC. Thus, article [5] applied the technology of porous silicon surface by etching the surface for nanostructures of the silicon samples for increasing both a life cycle and photoluminescence. This technology is actually cheap and can be used for SC. But the article replicates the experiments that were conducted with the front surfaces of SC less than 20 years ago [6]. There is only one exception – the nanostructures did not exist then.

A hybrid technology PS for the surface treatment was applied in [7]. Vanadium oxide was embedded in the structures of PS and led to a significant reduction in the reflectivity. Micro particles of PS were made from the mixture of SiO_2+αMg to be deposited at the surface of the substrate with the subsequent etching of the surface to obtain the required morphology in [8]. A multi-layered structure of PS was explored in paper [9]. Transport properties in this work were examined by using volt-ampere characteristics (VAC).

In space they use solar panels with multi-transition structures based on GaAs and the presence of concentrators; they can reachefficiency larger than 50 % [1], but are expensive.

Thin-film category of SC comprises a very small percentage of the total industrial production. In spite of its cheapness, the shortcoming is the high degradation of parameters of the thin-film SC, which limits their operation to 5–10 years.

In order to explain the properties of PS, researchers propsed a number of models that explain possible mechanisms for the formation of pores in the layers of PS. These models can be divided into several groups:

a) models that describe quantum limit of charge carriers in Si crystals of the nanometer size [10];

b) models that describe localized emission caused by the polysilanes of Si or hydrides, which are formed at the surface of PS during its growth as a result of the passivation of torn bonds along the surface [11];

c) models that describe the formation of a specific class of Si–O–H compounds (siloxanes) [12];

d) models that combine the theory of quantum limits of carriers and the existence of regions with local defects along the surface [13] the so-called hybrid models that better describe the optical properties of a porous film.

Authors categorized various models using different terms, such as "mathematical", "chemical" or "physical". In more recent papers, they were divided by dimensional porosity (micropores, mesopores, macropores), and universal models were also used [14].

If we assume that the process of pore formation can be described by one unified model that includes both the stage of nucleation and all stages of the growth of a pore, then this model would natuarally belong to a group of universal models. The aforementioned model was developed in article [15], in this case, at the bottom of the pore is a virtual passive film, which prevents direct contact between electrolyte and the substrate. All processes that occur at the boundary of silicon/fluid are fully accounted for. The model created, by using current oscillations on the volt-ampere characteristics, forms nano-, meso-, and macropores depending on the orientation of the crystal. An analysis of linear stability includes the models that combine transport phenomena of openings in a semiconductor, and the ions in electrolyte. The instability of planar surface and the development of small perturbations on it can be solved mathematically, for the processes of nucleation of pores at the surface of silicon. These models cannot be applied to explain the process of growth of pores in the required direction and this is their main shortcoming.

Etching ensemble of macropores on the surface of semiconductors and metals was presented based on the defect-deformational mechanism of spontaneous formation. It is based on the understanding of generation in the near-surface layer of the crystal that is exposed to etching, point defects (interstitials and vacancies). As a result of defect-deformational instability, there occurs a stationary hexagonal periodic defect-deformational structure. A nonlinear computer analysis of the film defect-deformational model [16] on an isotropic surface revealed that deep anode etching of pores at the second stage of etching proceeds on the clusters of vacancies that form a hexagonal cellular surface structure of the nucleation centers.

For a deeper understanding of the processes of anodization of silicon of the p-type conductivity, it was necessary to design a model of the etching reaction in a semiconductor. That is why a comprehensive model in article [17] was improved, originally devised for the n-type silicon, in order to explain the formation of macropores in silicon of the p-type conductivity. According to this model, the formation of pores is suppressed if the charge transfer is carried out by predominant thermo-electron processes, which is sensitive to the height of the barrier and is not sensitive to its width.

Calculation of electric field distribution near the bottom of a pore yields the model that is proposed in paper [18]; it also provides an explanation to the cause of local dissolution of silicon. The idea is postulated on that the growth of PS occurs when the two reactions compete. The first reaction is related to the formation of anode oxide and its boriding. In the second case, there is a formation of PS through direct dissolution of silicon in HF. The correlation between these reactions leads to the formation of PS or electric polishing of the surface.

They developed a model for silicon of the p-type that assumes that due to a small size of cystals in the walls between pores, there occurs a quantum limitation of the charge carriers, and, as a result, the rate of etching the walls slows. This model is such that it is now most often used to explain the formation of microporous structures, though, however, this model is not applicable to explain, for example, macroporous structures.

There is a model in which the formation of porous dielectric layer is caused by the action of mechanical stress on the process of pore formation or the saturation of surface layers with silicon vacancies [19].

Thus, the existing models and mechanisms of the formation of macropores in silicon cannot explain at present all of the experimental data. Even a widely used model from article [15], developed for the n-type silicon, cannot explain some of them. In addition, it seems more likely that the electrochemical process of the formation of macropores is of a more comprehensive nature. Therefore, a deeper understanding of the electrochemical processes in macropores that occur during the formation in a silicon substrate, requires more experimental data and their thorough investigation.

At present, humanity utilizes the so-called "Third generation" of photoelectricity, which is based on the quantum tubes and nanostructures. The use of silicon multifunctional porous materials in the SC structure will make it possible to control their conversion efficiency and reach the goal that would consistently improve the efficiency of conversion while reducing the cost of SC. Porous functional materials can be obtained in the form of layers with sufficiently large range of porosity, with the diameter of pores from nanometers to tens of micrometers [20].

It is necessary to conclude that most articles attempt to solve the problem on the reduction of reflectivity of the frontal surface while, at the same time, cutting the cost of technological process and improving the SC efficiency.

3. The aim and tasks of the study

The aim of present work is to create a multifunctional multitexture at the front surface of a photoelectric converter using hybrid technologies for obtaining the porous silicon. This will make it possible to create a multifunctional multitexture at the front surface of a photoelectric converter using hybrid technologies for obtaining the porous silicon.

To achieve the set aim, the following tasks had to be solved:

– to explore theoretically the relationship between the diameter of a pore d_p, porosity P and the region of specific surface S;

– to explore on the samples with high specific resistance the interaction between porosity P(t) and the region of specific surface S(t) of the PS grown by the electrochemical etching of silicon substrates;

– to examine by using the volt-ampere and spectral characteristics the effectiveness of SC conversion for the textures created by different technological methods.

4. Fabrication of multifunctional multitextures on the frontal surface of structures of photoelectric converters

4. 1. Modeling of an array of cylindrical pores to examine a relationship between the diameter of pore d_p, porosity P and the region of specific surface S

An important feature of porous layers is the large specific internal surface area (up to 1000 m²/cm³). The large surface area makes it a very active material; in this case, approximately 20 % of silicon atoms are arranged at the inner surface of PS [21, 22]. PS in terms of its morphology can be modeled as an array of cylindrical pores. This gives the possibility to theoretically investigate the relationship between diameter of a pore d_p, porosity P and the region of specific surface S. The array of cylindrical pores can be placed into the square (Fig. 1) or triangular (Fig. 2) lattices.

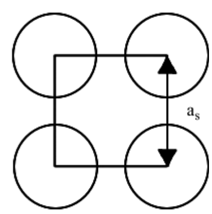

Fig. 1. Element of the array of cylindrical pores of PS, put in a square lattice

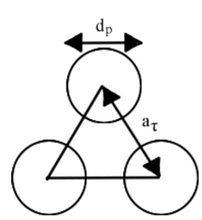

Fig. 2. Element of the array of cylindrical pores of PS, put in a triangular lattice

We can obtain for these two cases the dependences of the coefficient of porosity on the diameter of a cylindrical pore and specific surface S:

– for the square lattice

$$P = \frac{\pi}{4}\left(\frac{d_p}{a_s}\right)^2; \quad (1)$$

– for the triangular lattice

$$P = \frac{\pi}{2\sqrt{3}}\left(\frac{d_p}{a_T}\right)^2, \quad (2)$$

where d_P is the diameter of the pore and a_S, a_T are the dimensions of an elementary cell (Fig. 2, 3).

Considering the above equations, it is important to investigate when the pores start to merge. Given the above-mentioned equations, this will occur when $d_P=a_S$ and $d_P=a_T$. In this case, porosity in the case of square lattices takes values P>78.5 %, and in the case of triangular lattices – P>90.6 %. In both cases, the region of specific surface S is assigned by the same expression:

$$S=\frac{4P}{d_P}. \quad (3)$$

The PS formed on p-Si or n-Si typically has clear differences under conditions of the formation of pores by the size, orientation and degree of branching. In this case, the PS, formed in the dark or under lighting, has a difference in morphology. The PS, formed under the frontal lighting or lighting of the reverse side of the plate is also different by parameters. The most visible functional effect on the morphology under all conditions of its formation is exerted by the concentration of an alloying admixture. In particular, the size of pores depends on the type of an alloying admixture and its concentration. With increasing concentration of the alloying admixture for p-Si, the size of pores grows. In contrast, for n-Si, the size of pores decreases with its increasing concentration [23].

An increase in the diameter of a pore is directly associated with the period of etching. In this case, the surface area of the walls of the pore will increase. Over a certain period, this leads to an increase in the area of specific surface S(t) until the pores are combined. The region of specific surface will increase until porosity reaches the value of ≈78.5 % (for square lattices) or ≈90.6 % (for triangular lattices), and will decrease after the pores are combined.

4. 2. Results of examining the indicators of PEC with a multifunctional microtexture at the front surface

We examined the interaction between porosity P(t) and the region of specific surface S(t) of the PS grown by various methods of electrochemical etching of silicon substrates on the substrates that are used to fabricate SC (about 1.5 Ohm·cm).

The samples were chemically prepared in electrolytes using three different concentrations of hydrofluoric acid (35 % HF, 25 % HF and 15 % HF). The starting values of parameters were S(0)=334 m^2/cm^3, P(0)=68 % for the electrolyte with 35 % HF; S(0)=218 m^2/cm^3, P(0)=81 % for the electrolyte with 25 % of HF; and S(0)=137 m^2/cm^3, P(0)=91 % for the electrolyte with 15 % of HF. Results of the experiments indicate that the rate of loss of mass M(t) (Fig. 3) decreases with an increase in the duration of etching.

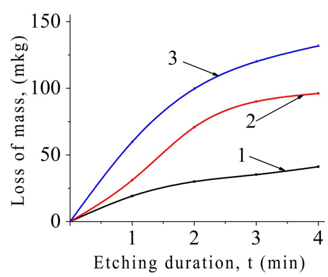

Fig. 3. Dependence of mass loss in the sample on the duration of etching for three etchants with amount of hydrofluoric acid: 1 – 15% HF; 2 – 25 % HF; 3 – 35 % HF

The basic principle in the formation of PS layers was the need to bring the process of their growth maximally close to the technology to create highly effective SC. The classic technology of the formation of PS by electrochemical method [24] was adopted as a basis for creating the SC with a microtexture.

For the growth of porous layers, we used both polished and textured KOH (100)-oriented silicon substrates of the p-type created by the method of zone melting with specific resistance 1.5 Ohm·cm, thickness 300 μm, polished by etching on both sides. The main stage was obtaining the emitter of the n+-type with thickness 0.4 μm and surface conductivity about 40 Ohm/cm^2. It was formed at 830 °C by the diffusion of phosphorus from a liquid source of $POCl_3$. Parameters of the technological process: 1 – 20-minute diffusion with subsequent 20-minute treatment in N_2; 2 – the next thermal treatment at 1050 °C. Key parameters of the process are the current density, the concentration of HF in electrolyte and specific resistance of the silicon substrate.

The growth of PS layers was conducted under galvanostatic mode with the use of teflon electrochemical cell whose design implied ohmic contact to the metallized backside of the silicon substrate. As the electrolyte, at anodizing, we employed the alcohol and aqueous-alcohol solution of hydrofluoric acid.

The formation of PS at the surface of emitter of the n+-p transition was carried out without the use of additional lighting and was enabled by the injection of openings of the base layer at direct displacement of the diode structure. At the first stage of this hybrid technology, chemical etching forms a crater-like macroporous texture. The second phase of electrochemical etching obtained kolonopodibna texture. A column-like texture may be controlled by both the depth and height of the received columns, adjusting the technological parameters of PS growth and the composition of the base acid solution.

Depending on the technological parameters, it is possible to form the layers of macro-, micro- or nanopores. Nanopores are formed by the relatively high values of HF concentration and density of the current. If the current and/or the concentration of HF are very low, then it is possible to obtain macropores. By changing the technological parameters, it is possible to receive a wide range of pores, from nano-PS to macro-PS. The electrolytes for experiments were prepared at the degree of concentration in hydrofluoric acid of 15 %; at some stages of the technological process, the current density was maintained at up to 76.4 mA/cm^2. The action period of different currents in the process of galvanostatic mode also varied, making it possible to cultivate layers different in thickness and porosity.

Using the proposed technology of obtaining the multi-functional multitexture based on PS, when receiving the macro-, micro- and nanotexture of PS in one technological process, we fabricated two laboratory samples of SC for comparing their output parameters.

The first (standard) was made using a simple pyramid texture. In the second, using the hybrid technology, we obtained a frontal microtexture. Parameters were measured by using the volt-ampere characteristics (VAC) with their conversion efficiency η defined. For the standard sample of SC, conversion efficiency η~12.6 %, and for the sample with a front microtexture – conversion efficiency is η~16.4 %. VAC were measured by the spectral conditions of AM 1.5G, experiments for both samples of SC were carried out on the silicon substrates of the same parameters and area.

The main parameters of the SC structure (Fig. 4) with a frontal microtexture were determined by using a specialized unit for the measurement and calculation of the volt-ampere characteristics (VAC), original electrical characteristics andefficiency of SC. It uses a simulator of the solar light for a spectrum of AM 1.5G. Results of the measurements were processed by a specialized siftware and were represented in the form of VAC of the photoconverter indicating the maximum values of photocurrent and output voltage, short circuit current and the voltage of idling, as well as the coefficient of filling and SCefficiency (Fig. 4).

Fig. 4 shows example of the diagram (print-out) of volt-ampere characteristics, produced by the software of specialized unit for the measurement and calculation of VAC. The sample of PEC was fabricated on a silicon substrate with a multitextured surface without ARC. Specialized program, along with SC, indicates the calculated VAC parameters: maximum values of photocurrent and output voltage, short circuit current, voltage non-working course, fill factor, efficiency.

Further improvement in the conversion efficiency can be achieved by controlling the specific resistance of silicon wafers, length of the diffusion of carrier and by the application of process of passivation of emitter and local diffusion of the reverse surface. Maximal characteristics can be attained by the additional application of ARC on the surface of a microtexture. It is estimated to increase conversion efficiency of SC by not less than ~18 %.

Fig. 5 shows spectral characteristics of reflectivity, measured for the surfaces fabricated by chemical texturization and for multifunctional multitextures. For the comparison, there are spectra of reflectivity for the textures of chaotic pyramids and a polished surface.

Thus, the impact of these two different layers in a multitexture that are defined by the diameter of a pore is indistinguishable. In order to obtain double-layered PSon n-Si, conditions must be controlled in a strictly defined fashion. The pores, formed under the influence of the layer of spatial charge, and the pores, formed under the influence of the resistive layer, should have a small difference in size. That is, if the sizes of the pores are within the same order, this can be used.

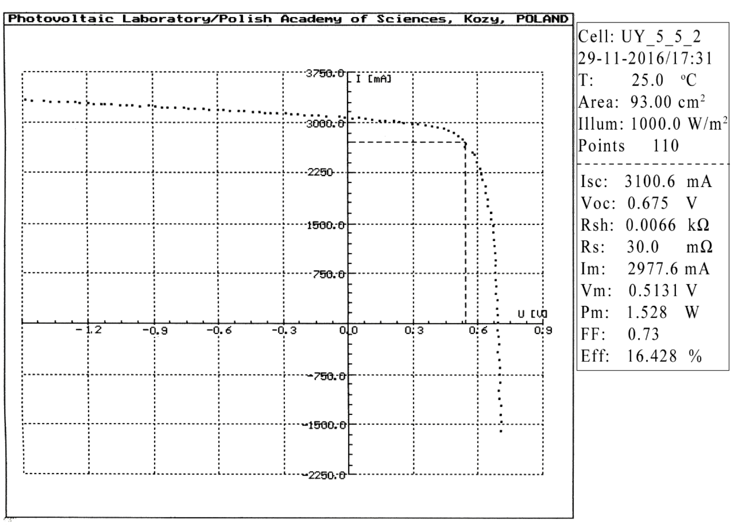

Fig. 4. Diagram (print-out) of the volt-ampere characteristics produced by the software of specialized unit for the measurement and calculation of SC VAC, with indicated maximum values of photocurrent and output voltage, short circuit current and the voltage of idling as well as the coefficient of filling and SC PE

When analyzing spectral characteristics in the range of 400–1150 nm, it should be noted that the creation of a multilayer structure of ARC on the multitexture will significantly reduce the integral reflectivity coefficient of the frontal surface of SC and improve efficiency.

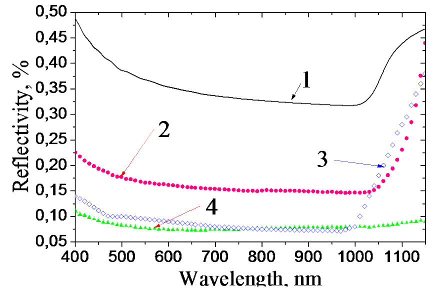

Fig. 5. Spectral characteristics in the range of 400—1150 nm: 1 — polished surface of Si; 2 — chemical texturizing; 3 — chaotic pyramids; 4 — microtexture without the application of ARC

5. Discussion of results of examining the fabrication of PEC with a multifunctional multitexture

PS is characterised by the multifunctionality of properties and very specific morphological features, the process of its formation is a multifactorial one. A number of theories describe different aspects of the formation of PS. The theories of the formation of PS considering the localization of carriers on the tip of pores in the process of its growth, impact of the diffusion of openings on the intensity of pore growth, effect of the chemical activation of a surface and the interface between PS and the substrate.

Intensive research into the inner surface led to another point of view on the future PS use, which may depend on its large specific surface. For this purpose, it would be desirable to fill the free space of PS with some fillers or PS of another dimensionality, which does actually form the multitexture. If most of the volume of pores is filled with applicable fillers or PS of another dimensionality, then there is the need in the large sizes in pores of the material, for example, macro-PS, because the size of the pores will determine the efficiency of their filling will filler. The size of the pores and the parameters of the region of specific surface of the frontal side of a solar element (SC) will together affect efficiency of the photoelectric conversion.

Experiments for both samples of SC were conducted on the silicon substrates with the same parameters and area. An analysis of VAC of the two fabricated samples indicates a significant increase (by 30 %) in the conversion efficiency η for the sample of SC that employs microtexture. The very efficiency η of the conversion increased by 3.8 %.

If we functionally change the current density along the surface of a pore, then it is possible to create at the frontal surface PSwith pores of various shapes. In this case, significant difference is demonstrated by the PS, formed in the dark or under lighting, as well as PS formed on the p-Si or n-Si substrates. It has distinct differences under conditions of the formation of the size of a pore, the orientation and degree of branching. PS, formed under frontal lighting or lighting from the reverse side of the plate is also different by these parameters. Among all the conditions for the formation of PS with pores of various shapes, the most pronounced functional effect is displayed by the concentration of an alloying admixture. In particular, the size of pores depends on the type of substrate's alloying admixture and its concentration, while the size of pores usually increases with increasing concentration of an alloying admixture for p–Si, but decreases with the concentration of an alloying admixture for n–Si. In terms of quantity, the size of pores is the most used parameter to characterize the size of the element along with the physical and chemical properties of PS. It should be noted that the spectral characteristics for the multitexture in the range of 400–1150 nm has a significant feature. It practically has no high values in the infrared range, in contrast to all other examples. This yields the integral coefficient of reflectivity for the frontal surface of multitexture, which is significantly lower ~7 % than other integral coefficients. For the chemical texture ~17.5 %, random pyramids ~11.2 %, for the polished surface of Si – larger than 35 %.

6. Conclusions

1. PS was modeled as an array of cylindrical pores located in the square or triangular lattices. Important is the moment when the pores start to merge. In this case, porosity in the case of square lattices will reach P>78.5 %, and in the case of triangular lattices – P>90.6 %.

2. Depending on the technological parameters, it is possible to form the layers of macro-, micro- or nanopores. At relatively high values of the concentration of HF and current density, it is possible to form nanopores. Electrolytes for the experiments were prepared at the concentration degree of hydrofluoric acid at 15 %; in this case, at some stages of the technological process, the current density was maintained up to 76.4 mA/cm^2.

3. We fabricated 2 samples of SC for the experiments. Conversion efficiency for the sample with a frontal microtexture made by using the hybrid technology without the use of ARC reached η~16.4 %. Maximal characteristics can be obtained by the additional application of ARC on the surface of microtexture. It is estimated to increase the conversion efficiency of SC by not less than η~18 %.

Intensive research into the inner surface led to another point of view on the future PS use, which may depend on its large specific surface. For this purpose, it would be desirable to fill the free space of PS with some fillers or PS of another dimensionality, which actually forms the multitexture. The size of the pores and the parameters of the region of specific surface of the frontal surface of a solar element (SC) will together influence improvement in the efficiency of photoelectric conversion.

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