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АНАЛИЗ ОСОБЕННОСТЕЙ ВОЗНИКНОВЕНИЯ ПОТЕНЦИАЛОВ В МНОГОКОМПОНЕНТНЫХ КЕРАМИЧЕСКИХ КОМПОЗИТАХ НА ОСНОВЕ ТУГОПЛАВКИХ БЕЗКИСЛОРОДНЫХ СОЕДИНЕНИЙ (ЧАСТЬ 2)

Доказано, что для получения максимального значения коэффициента термо-э. д. с. металлические включения должны иметь вытянутую цилиндрическую и объемную форму. Порог перколяции должен быть около 2, а фрактальная размерность кластера должна принимать значение около 2,4. Полученные значения противоречат существующей теории возникновения термо-э. д. с. в многокомпонентных композитах и требуют дальнейших исследований.

Ключевые слова: теория перколяции, теория Скала, термо-э. д. с., карбид гафния, коэффициент термо-э. д. с., металлические включения.

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DEVELOPMENT OF EFFICIENCY IMPROVEMENT METHOD OF PHOTOVOLTAIC CONVERTERS BY NANOSTRUCTURIZATION OF SILICON WAFERS

Визначено технологічні режими отримання шару поруватого кремнію. Поруватий кремній доцільно формувати методом електрохімічного травлення у розчині плавикової кислоти. Встановлено, що товщина пористого шару корелює з часом травлення. Пористість демонструє майже лінійну залежність від щільності струму. Визначено доцільність використання пористого кремнію у якості основного матеріалу для виготовлення фотоелектричних перетворювачів (ФЕП).

Ключові слова: *фотоелектричні перетворювачі, пористий кремній, коефіцієнт відбиття, електрохімічне травлення, наноструктури.*

1. Introduction

Modern social, environmental and economic trends of society development determine the key role of energetics in transition the strategy of society transition to sustainable development. Energy issues serve as key global issues of the day, the nature of which decision directly determines not only overcoming the ecological crisis, but construction of the global economy and development strategies. Hence, the urgency to find alternative ways to provide human by energy is followed.

The growth of the industrial capacity of mankind is based mainly on the progress in the field of energy technologies. Energetics plays a central role, as in the emergence of ecological crisis and to overcome it. Most important mechanism for harmonization of «society-energy-biosphere» system is a gradual transition to large-scale implementation of alternative energy sources in the energetics.

The most promising method of alternative energy is photovoltaic solar energy conversion method through existing advantages:

1) direct energy conversion of light photons into electricity;

2) diversity of basic framework for the production of solar cells;

3) ability to create modular systems of different capacities;

4) use of concentrated solar radiation;
5) quietness;
6) ease of use;
7) environmental friendliness and so on.

Disadvantages of using solar energy often are the following:

1) high cost of solar panels;
2) electricity generation only during daylight hours;
3) depending on the climatic conditions;
4) need for large areas to photovoltaic stations;
5) problems of energy storage;
6) imperfect technology, low efficiency and so on.

Imperfect creation technology of photovoltaic converters (PVC) and their low efficiency is a major deterrent of global replacement of traditional energy to renewable. So, there is a need to develop innovative technologies that can increase efficiency and other electrical characteristics of solar panels.

In this regard, relevant studies are devoted to finding ways to improve the processes of manufacturing photovoltaic converters. In the development of appropriate technological solutions it should be taking into account characteristics of the materials used as raw material for PVC.

2. The object of research and its technological audit

The object of this research is the process of preparing silicon wafers for further use as the main component of solar panels.

One of the problems in this process is the degradation of silicon in service and high rates of reflection coefficient, which greatly affects the PVC performance.

Technological audit was conducted to identify the basic laws of state impact of silicon wafers on PVCs technology performance. Its aim is to determine the feasibility of nanostructurization of single-crystal ingots and passivation of their surface.

The basis of the most common commercial photovoltaic devices is solid single-crystal silicon solar cells with *p-n*-junctions [1]. Well established technology for producing and processing of single-crystal silicon allows maintain key positions for solar cells based on it in the near future [2]. The high cost of technologies for single-crystal silicon films and multicomponent technology of multilayer semiconductor structures are prevented to widespread PVCs use. Mono-Si-based solar cells have efficiency is about 15–20 %.

Because of a high refractive index of silicon ($n = 3{,}5$), much of the solar radiation is reflected from the surface of the photovoltaic converter (value can be ~35 %) and therefore do not contribute to the process of generation of electron-hole pairs. This leads to a decrease in the efficiency of converters [3]. Typically, this problem is solved by application of antireflection coatings on the surface of solar cells. The use of such coatings leads to the increase of the conversion efficiency, extend the life and improve electrophysical and operational performances of photovoltaic converters.

The technical and economic alternative to existing devices in the near future can make the items, which operation is based on photoelectrochemical methods of surface conversion.

3. The aim and objectives of research

The aim of this research is to develop technological solutions for treatment of the front surface of the silicon solar cells and to establish feasibility of porous layer formation on their surface.

To achieve this aim it is necessary:

1. Identify technological modes of obtaining porous silicon layer.
2. Investigate the correlation between the parameters of porous layer and conditions of crystal etching.
3. Set the feasibility of using porous silicon as the main material for PVCs.

4. Literature review

Among areas to overcome the problem of PVC imperfection it can be allocated the next:

– Search of new materials for solar panels [4, 5].
– Preparation improvement of elementary base [6–9].
– Use of antireflection coatings for PVC [10, 11].

In particular, [4] proposes the photoelectric converter having a two-way sensitivity and made on based crystals as *p*- and *n*-type. Reduction of the rate of surface recombination in these PVCs is ensured in the implementation of techniques that specific to PVCs with PERL-structure. Efficient absorption of solar radiation is provided by using chaotic textures of direct pyramids type without further photolithography. The reflection coefficient from the front PVC surface with DSBC-structure is 5,8 %. Such PVCs in exposure mode AM1,5 have the following output parameters: U_{hh} – 704–706 mV, J_{sc} – 42,2 mA/cm^2, FF – 0,82–0,83. Also their technological aspects allowed to achieve the following values of output parameters: U_{hh} – 663–665 mV, J_{sc} – 32,6–36,6 mA/cm^2, FF – 0,76–0,78, η – 16,8–18,6 %.

The authors of [5] proposed resource-saving industrial technology of radiation-stable PVCs of $n+$-p-$p+$ type with area of 7 cm^2 on the basis of low-cost single-crystal silicon KDB-10 grown by the Czochralski method, with thick $t_k = 350 \pm 50$ microns, orientation (111) and (100), which in exposure mode AM0 have efficiency of 12–15 %. Development of such PVCs provided an opportunity to use them to build solar cell of spacecraft KS5MF2 «Micron» – the first in Ukraine from spacecraft series «Microsatellite».

The technology of porous layers of semiconductors by electrochemical etching is represented in [6–8]. The use of porous silicon for solar panels was investigated by the authors of [9]. It was proved that the increase in conversion efficiency (25–30 %) is achieved for the por-Si solar cells compared to cells without porous layer.

The authors of [10, 11] believe that the most promising antireflection coating for photoelectrochemical cells is zinc oxide (ZnO). This material is cheap, accessible and non-toxic. But consensus on the morphology of these coatings is still not there.

Thus, the results suggest that the consensus on the possibility of improving the efficiency of photovoltaic energy conversion does not exist. Obviously, the choice of the most appropriate technical solution should be based on experimental and theoretical studies of the structure and properties of porous silicon layers.

5. Materials and methods of research

Samples of single-crystal n-type silicon of solar grade (silicon with silicon content of more than 99,99 % by weight, with an average lifetime of non-equilibrium carriers of 25 microseconds and electrical resistivity of 10 Ohm*cm) were selected for experiment. Surface orientation: (111) (100) (001). Silicon single crystals were grown by the Czochralski method and doped with phosphorus. Then the ingots were cut into 1 mm thick wafers by the method of wire cutting by the diamond-impregnated wire. Samples were purified in acetone and isopropanol before thee experiment.

The layers of porous silicon were formed by electrochemical etching in a solution of hydrofluoric acid at room temperature. This method is the most common for making porous semiconductors, due to the simplicity and low cost [6]. In addition, this method is different from other by rate and accountability of the process. Multivariate implementation of the method is technically possible, combining it with other additional (external) physical and technological influences, factors, fields and so on.

Another factor that makes electrochemical crystal processing as attractive method for forming porous spaces is the possibility to adjust the pore size from a few nanometers to tens of micrometers [7]. This is achieved by adjusting the doping level and orientation of the surface wafers and etching conditions. In addition, a wide range of porous characteristics such as thickness, porosity, effective area surface and morphology may control, changing etching conditions.

The conditions of the experiment were chosen in the following range of characteristics:

a) current density – from 10 mA/cm^2 to 2,2 A/cm^2;

b) etching time – from 5 to 30 minutes;

c) electrolyte – $HF:H_2O = 1:1$;

d) electrolyte temperature – 20 °C.

The scheme of the simplest galvanic cell is shown in Fig. 1. The silicon wafer serves as the anode, the cathode is platinum.

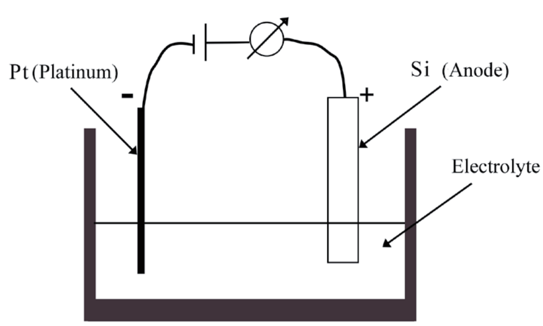

Fig. 1. The scheme of device for electrochemical silicon etching

Samples after experiment had several stages of cleaning: washing in ethanol, deionized air drying and annealing in a stream of nitrogen. Annealing is carried out to stabilize the properties of porous wafers. Oxide layer is removed from semiconductor surface using this passivation method. Thin crystalline film of chemically inert material is formed instead of this layer. Such film can do the properties of ultrathin buffer layer and protect the semiconductor surface from contact with aggressive components of the environment.

6. Research results

The thickness of the porous silicon layer on the Si substrate adjusts with etching time. The porosity, i. e. the proportion of voids in the porous layer, is depended on the current density of the electrolyte, resistance and Si substrate doping density.

Anodic reaction during the formation of the porous layer can be written as:

$$Si + 6HF \rightarrow H_2SiF_6 + H_2 + 2H^+ + 2e^-.$$

Atoms of silicon creates adatoms SiF_6^{2-}, requiring the presence of ions F^- (from HF solution) [5]. Adatoms escape from the wafer surface and create rudimentary positively charged holes. The concentration of holes in positively charged silicon is enough (about 10^{14}–10^{18} cm^{-3}) for the formation of nanometer-sized pores.

When using a fairly low current density (from 10 to 200 mA/cm^2), there is a local solution of single-crystal silicon surface. Rudimentary pores are formed in places with surface defects, further the pores are growth into the depth of the silicon substrate. This process occurs by diffusion of the holes to the interface Si-electrolyte.

In the case of high current density (0,3–1 A/cm^2), when the number of holes moving to the border Si-electrolyte is very high, is the most appropriate is etching of upper regions of the silicon substrate. It provides uniform etching of silicon surface and formation of a smooth substrate surface (called electrolytic polishing process).

Increase of current density above the critical point at the end of anodizing process leads to disconnection of the porous silicon film from Si substrate. Such behavior at high current density is useful for separating porous layers from substrate.

Fig. 2 shows the morphology of porous silicon grown on the surface of *n*-type mono-Si. The porous layer has a developed structure, pore size ranging from 0,2 to 3 microns, porosity is about 70 %. The thickness of the porous layer is 35 microns. Pores have a columnar structure.

In this case, there is heterogeneity of porosity and thickness of the porous layers. Most likely, this is due to the presence of bubbles that formed in the electrolyte and bonded to the silicon surface. To avoid this heterogeneity, concentration of HF should be locally constant on the surface of the silicon substrate.

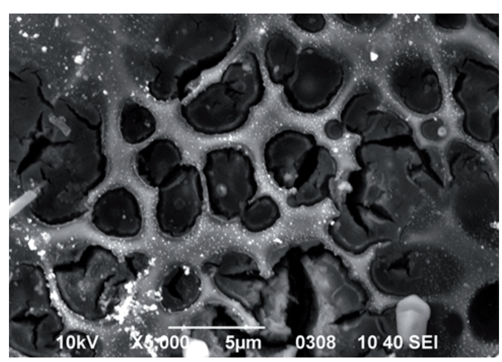

Fig. 2. Morphology of por-Si (100): current density j = 150 mA/cm^2, etching time t = 20 min, electrolyte $HF:H_2O = 1:1$

Removal of the bubbles from the surface of a silicon wafer, and, thus, obtaining of homogeneous porous silicon layers is done by means of electrolyte mixing. The distance between the silicon wafer and the platinum cathode also affects the uniformity, while form of the platinum cathode virtually no affects homogeneity.

The thickness of the porous layer depends primarily on etching time. Fig. 3 shows the experimental dependence of the thickness of the porous silicon area (100) under the following etching conditions: current density $j = 150$ mA/cm^2, etching time $t = 20$ min, electrolyte $HF:H_2O = 1:1$. It is shown that the average growth kinetics of porous silicon layer is about 2,2 m/min. It is noted that this parameter is determined for each etching case separately.

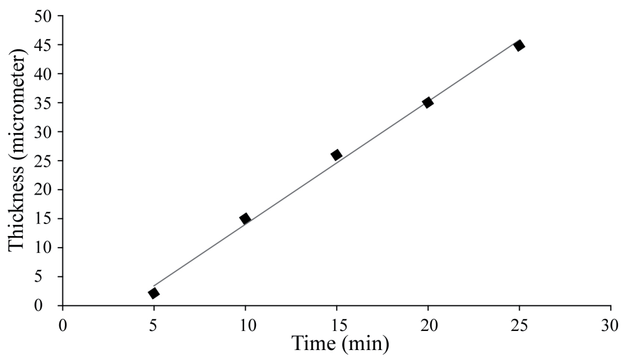

Fig. 3. Increase of the porous silicon layer thickness (100) during electrolytic processing in 50 % solution of hydrofluoric acid for 20 minutes at a constant current density $j = 150$ mA/cm^2

The porosity shows a nearly linear dependence on the current density in the range of 30 to 200 mA/cm^2 (Fig. 4) at constant other parameters.

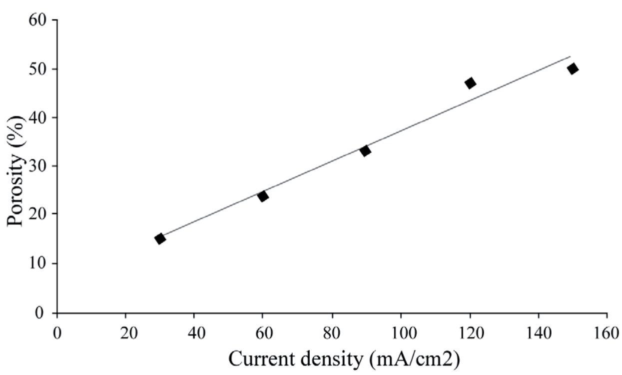

Fig. 4. Porosity dependence on the current density (for por-Si (100), current density $j = 150$ mA/cm^2, etching time $t = 20$ min, electrolyte $HF:H_2O = 1:1$)

It should be noted that porous silicon is a special form of crystalline silicon. The crystal structure of porous silicon is a network of silicon in nanoscale areas surrounded by empty space with a very high ratio of surface area to volume (up to 10^3 m^2/cm^3) — so-called effective area of the crystal. Quantum effects in this case play a fundamental role. In particular, the photoluminescence of porous silicon shows shear in short-wave spectrum in relation to the crystal.

Materials used as raw materials for the manufacture of solar cells have a number of requirements, including:

1) direct band gap structure;
2) band gap width from 1,1 to 1,7 eV;
3) must be composed of readily available and non-toxic materials;
4) good photovoltaic conversion efficiency;
5) long-term stability.

Silicon is the second most abundant element in the earth's crust (35 %) after oxygen. This is the basic material for photovoltaic conversion of solar spectrum radiation ranging from ultraviolet to near-infrared region of the spectrum, but it can absorb a small portion of solar radiation that can convert photons with energy of band gap of silicon. The theoretical curve for the conversion efficiency of solar cell material compared to the band gap shows that silicon (1,1 eV) is not at the maximum of the curve (about 1,4–1,5 eV), but relatively close to it. Efficiency for ideal silicon solar cells can reach about 30 % (at 300 K) [5].

Photoelectron properties associated with non-direct band gap structure and high reflectivity of crystalline silicon (about 30–35 %) still do it as contender for making solar cells with high conversion efficiency. The high refractive index of crystalline silicon (about 3,5) in the solar spectrum (300–1100 nm) creates large optical losses that can be reduced by using antireflection coatings (ARC). Although availability of highly-effective double and triple antireflection coatings, most industrial crystalline silicon solar cells use simple and inexpensive single-layer ARC with relatively poor antireflection properties.

Opportunities for reflectivity minimization (by catching the light in the pores), an increase band gap width of porous layer of silicon (through quantum maintenance of the charges in microcrystallites por-Si) by changing the porosity allow to use the layers of porous silicon as anti-reflection coating, and as broadband photosensitive layer.

The high degree of surface roughness of the porous silicon allows for its use as ARC, because the textured surface reduces light reflection. In addition, scattering in por-Si may be due to the roughness relative to the thickness of the porous layer.

7. SWOT analysis of research results

Strengths. Among the strengths of this research it should be noted results in optimal conditions of silicon etching to formation of porous layers on its surface, as well as determining por-Si parameters from anodizing modes. In favor of this assertion are evidenced the above analysis results of contemporary world literature, where no such correlation. This is holding back the beginning of the use of porous silicon as the main base of the solar panels. Use of the proposed dependencies will optimize the electrochemical crystal etching process and produce the layers of porous silicon with strictly controlled properties.

Weaknesses. Weaknesses of this research are related to the fact that the proposed solution will be effective when using ultrapure dislocation-free silicon. Using polycrystalline wafer, electrochemical treatment will not lead to the formation of the porous layer. Wafers of monocrystalline silicon are known to be much more expensive than other versions of this semiconductor. This can lead to an increase in the cost of silicon solar panels. However, the expected significant increase in the efficiency of photovoltaic converters will minimize this factor.

Opportunities. Additional features that achieve the aim of research appear likely in these external factors. Considered technological object is widely studied by foreign scientists. Green energy program is introduced in the world. In Japan, there are programs «Sunshine» and «Moonlight». Germany has a government program that provides tax incentives to manufacturers of solar panels, which are mounted on rooftops. At the same time for several years there is a program «One hundred thousand solar roofs». A similar program «Million Solar Roofs» is in the US. Solar energy market is growing rapidly. Thus, the demand for solar cells of new generation will grow. This is a motivating factor for further research in this area.

Threats. Difficulties in the implementation of research results are related to the following factors. The first of these is knowledge-base of proposed technologies that require, on the one hand, the training of qualified personnel and, on the other hand, considerable resources. Another factor is the market of modern solar cells. Leaders of industrial solar cells today are Chinese companies that offer solar panels are much cheaper than other manufacturers. However, such PVCs are much inferior to their performance. It is necessary shift in the direction of product quality from the price.

Thus, SWOT-analysis of research results allows to determine the main directions for the successful achievement of the objectives of research, including: further improvement of obtaining porous layers of semiconductors, integration of porous layers in elementary base of photoelectrochemical converters, investigation of surface passivation modes for the purpose of refusal of ARCs.

8. Conclusions

1. The technological modes of obtaining porous silicon layer are determined. The porous silicon is expedient form by electrochemical etching in a solution of hydrofluoric acid. It is established that nitrogen annealing of the samples after etching leads to stabilization of the properties of porous silicon layer.

2. The dependence between the parameters of porous layer and conditions of crystal etching is investigated. It is established that the thickness of the porous layer is correlated with the etching time. The porosity shows a nearly linear dependence on the current density in the range of 30 to 200 mA/cm^2 at constant other parameters.

3. The feasibility of using porous silicon as the main material for making PVCs is established. The high degree of surface roughness of the porous silicon allows for its use as ARC, because the textured surface reduces light reflection.

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РАЗРАБОТКА СПОСОБА ПОВЫШЕНИЯ ЭФФЕКТИВНОСТИ ФОТОЭЛЕКТРИЧЕСКИХ ПРЕОБРАЗОВАТЕЛЕЙ ПУТЕМ НАНОСТРУКТУРИРОВАНИЯ КРЕМНИЕВЫХ ПЛАСТИН

Определены технологические режимы получения слоя пористого кремния. Пористый кремний целесообразно формировать методом электрохимического травления в растворе плавиковой кислоты. Установлено, что толщина пористого слоя коррелирует со временем травления. Пористость демонстрирует почти линейную зависимость от плотности тока. Определена целесообразность использования пористого кремния в качестве основного материала для изготовления фотоэлектрического преобразователя (ФЭП).

Ключевые слова: фотоэлектрические преобразователи, пористый кремний, коэффициент отражения, электрохимическое травление, наноструктуры.

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