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Transparent Conductive Oxide Layers and Their Application in Solar Energetic

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Solar cells fabricated on the basis of semiconductor-insulator-semiconductor (SIS) structures are promising for solar energy conversion due to their relatively low cost. These structures are obtained by deposition of transparent conductive oxide ITO films onto different crystal substrates.

The ITO layer is deposited by spraying alcoholic solution of $InCl_3$ and $SnCl_4$. Films with conductivity 4.7 10^3 $Ohm^{-1}.cm^{-1}$ and transmission coefficient 87 % were obtained. For the fabrication of Si and InP based SIS structures respective wafers with the concentration 10^{15} cm^{-3}-10^{17} cm^{-3} were used as substrates. For ITO-nSi solar cells with area of 8.1 cm^2 in AM1.5 conditions the efficiency is 9.57 %, for ITO-pInP solar cells the efficiency reaches 11 %. Bifacial ITO-nSi solar cells have been also fabricated. The quantum efficiency (QY) was studied at frontal and back illumination. In the region of wavelengths from 400 nm to 870 nm the value of QY changes in the limits 0.60 – 1 in the case of frontal illumination. For back illumination QY is equal to 0.20–0.33 in the same region of the spectrum. The photoelectric parameters of the ITO-nSi bifacial solar cell in AM 1.5 conditions are: for the frontal side U_{oc}= 0.425V, I_{sc}= 32.63 mA/cm^2, FF = 68.29 %, Eff.= 9.47 %, for the rear side U_{oc}= 0.392 V, I_{sc}= 13.23 mA/cm^2, FF = 69.28 %, Eff.= 3.60 %.

Key words: bifacial, ITO, silicon, solar cell, TCO.

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Introduction

Thin layers of transparent and conductive oxides (TCO) with high optical transparency and electrical conductivity due to their very interesting properties attract the attention of a large number of researchers and have found a wide range of applications in science and technology. These materials can serve as an excellent protective layer and are used in different applications, such as electrode material for spectroscopic studies, and protective large band windows for different substrates. Their ability to reflect thermal infrared heat is exploited to make energy conserving windows. Electrical current is passed through TCO layers to defrost windows in vehicles; they dissipate static electricity from the windows of xerographic copiers [1], [2]. TCO layers have been obtained from a wide variety of materials – oxides of tin, indium, zinc, cadmium and other metals. In solar cells fabrication the most utilized materials are tin oxide, indium oxide and their mixture known as indium tin oxide (ITO). A large application of TCO materials is found in photovoltaic (PV) conversion of solar energy. Thin layers of ITO have been deposited onto different semiconductors to obtain what is often called Schottky barrier - like heterojunctions or semiconductor-insulator-semiconductor (SIS) structures [3]-[11]. The high electrical conductivity of ITO is exploited in front surface electrodes for solar cells. In this case the ITO layers are also antireflection coatings.

In this communication, deposition and properties of ITO layers obtained by spray pyrolysis technique have been investigated. These layers, which are deposited on the surface of silicon and indium phosphide crystals, are used for obtaining SIS structures and respective solar cells on their base. The elaborated method is more simple and lower in cost in comparison with the usual technology of solar cells fabrication.

I. Deposition of ITO layers by spray pyrolisis technique

There are several different methods of obtaining TCO layers: magnetron sputtering, spray pyrolysis, chemical vapor deposition (CVD), et al. [2].

The spray pyrolysis technique is a simple and low cost method, whereas magnetron sputtering and CVD methods tend to involve the use of sophistical reactors and expensive vacuum installations. Therefore we have chosen the spray pyrolysis method for obtaining ITO thin films with a high conductivity and a major transparency in the visible region of solar radiation spectra. The ITO layers are deposited on the nSi crystals surface using the specially designed installation which contains four main units: the spraying system, the system of displacement and rotation of the support on which the substrate is fixed, the system of heating the substrate and the system of the evacuation of the residual products of the pyrolyse [12]. The heating system consists of an electric furnace and a device for automatic regulation of the substrate temperature (Fig. 1).

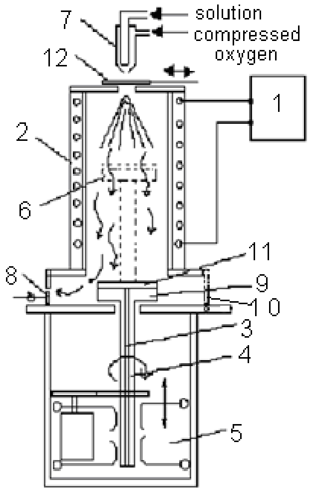

Fig. 1. Schematic image of the installation for ITO thin films deposition.
1-power unit; 2-electric furnace; 3-thermocouple; 4-rotating basis; 5-mechanism of basis rotation and moving; 6-basis position during layer reception; 7-system of spraying; 8-system of exhaust ventilation; 9-basis position during loading of silicon wafer; 10-cover; 11-silicon wafer; 12-shielding plate.

The silicon wafers are located on the support and with the aid of the displacement mechanism are moved in the deposition zone of the electric furnace. The construction of this mechanism provides the rotation of the support with the velocity of 60 rotations per minute, the speed necessary for the obtaining of thin films with uniform thickness on the all wafer surface. The alcoholic solution of the mixture $SnCl_4 + InCl_3$ is sprayed with the aid of compressed oxygen into the stove on the silicon wafer substrate, where the ITO thin film is formed due to thermal decomposition of the solution and the oxidation reaction. On the heated up substrate there are the following chemical reactions:

$$4InCl_3+3O_2 = 2In_2O_3+6Cl_2$$
$$SnCl_4+O_2 = SnO_2+2Cl_2.$$

The X-ray analysis showed that the ITO thin films had a cubic crystalline structure with a lattice constant of 10.14 Å. ITO films with maximum conductivity $4.7\cdot10^3$ $Om^{-1}cm^{-1}$ and maximum transmission coefficient in the visible range of the spectrum (87 %) were obtained from solutions containing 90 % $InCl_3$ and 10 % $SnCl_4$ at the substrate temperature 450°C, deposition rate 100 Å/min, spraying time 45 s.

II. ITO- nSi Solar cells

The solar cell cost reduction must be obtained by utilization of low cost fabrication technologies. An example of such technology is the spraying method, which permits the attainment of semiconductor-insulator-semiconductor (SIS) structures. The advantage of this technology is the elimination of the high temperature diffusion process from the technological chain and the utilization of low temperatures, no more than 500^0C in the fabrication process. A degenerated high conductive and high transparent oxide thin film (ITO) is used for the obtaining of the frontal layer, which is at the same time an antireflection coating and collecting electrode. The separation of light generated carriers is achieved by a space charge region which is in the basic semiconductor near the insulator layer. The possibility of such solar cells (SC) fabrication by a simple pyrolytic spraying method was shown in [3]-[11] where a mixture of indium and tin oxides (ITO) has been used.

New semiconductor materials are proposed for the fabrication of solar cells, but crystalline silicon continues to hold the dominant position in PV cell production. For the fabrication of SIS structures nSi wafers oriented in the (100) plane with resistivity 1.0 Ohm.cm and 4.5 Ohm.cm (concentrations $5\cdot10^{15}$ cm^{-3} and $1\cdot10^{15}$ cm^{-3}) and pInP crystals oriented in (111) A, (111) B and (100) plane with the carrier concentrations $p = (3...30)\cdot10^{16}$ cm^{-3}, were used. Insulator layers were obtained on the wafers surface by different methods: anodic, thermal or chemical oxidation. The best results have been obtained at the utilization of the two last methods. The chemical oxidation of the silicon surface was realized by immersing the silicon wafer into the concentrated nitric acid for 15 seconds. A tunnel transparent for minority carriers insulator layers at the ITO-Si interface have been obtained thermally, if the deposition occurs in an oxygen containing atmosphere. Ellipsometrical measurement showed that the thickness of the SiO_2 insulator layer varies from 30 Å to 60 Å. In the case of InP crystals a

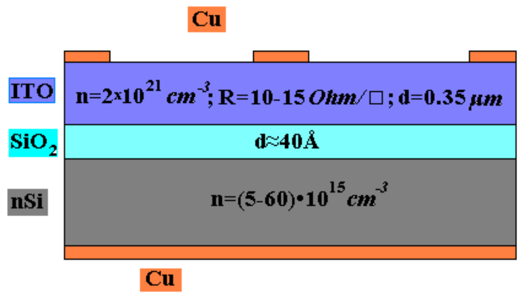

Fig. 2. Schematical appearance of Cu-ITO-SiO_2-nSi-Cu structure.

thin insulator P_2O_5 layer with the thickness 3-4 nm was formed on InP wafer surface during the ITO layers deposition. Ohmic contacts to pInP were obtained by thermal vacuum evaporation of 95 % Ag and 5 % Zn alloy on the previously polished rear surface of the wafer. The frontal grid was obtained by Cu vacuum evaporation. The investigation of the electrical properties of the obtained SIS structures demonstrates that these insulator layers are tunnel transparent for the current carriers.

In such a way obtained ITO- nSi and ITO- pInP SIS structures represent asymmetrical doped barrier structures in which the wide band gap oxide semiconductor plays the role of the transparent metal.

The schematic view of one of these structures is presented in Fig. 2. The obtained SIS structures were used for the fabrication of photovoltaic converters.

The spectral distribution of the quantum efficiency (QY) of the n^+ITO-SiO_2-nSi structure is presented in Fig. 3 (curve 1).

It is observed that in the region of wavelengths from 400 nm to 870 nm the value of QY changes on the limits 0.6-0.97. The photosensitivity measured in *A/W* for a fixed wavelength is determined from the ratio of I_{sc} and the incident light energy. The curve 2, Fig.3 represents the spectral distribution of the photosensitivity for the same sample. The photosensitivity is observed in the wavelength range of 350 nm -1200 nm and in the wavelength range of 485 nm -920 nm its value is more than 0.45 A/W.

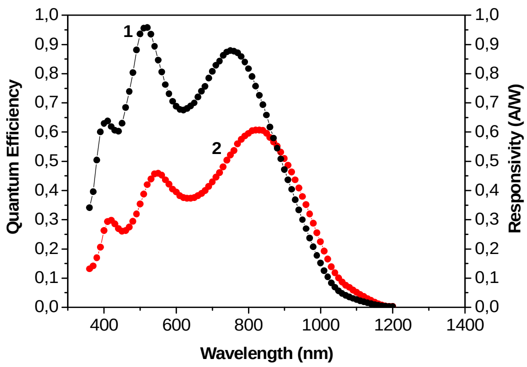

Fig. 3. Spectral distribution of the quantum efficiency (1) and photosensibility (2) of the n^+ITO-SiO_2-nSi solar cells.

For solar cells with active area of 8.1 cm^2 at the AM 1.5 illumination conditions the following photoelectric parameters were obtained: the short circuit current 25.11 mA/cm^2, the open circuit voltage 536 mV, the fill factor 71.14 %, the efficiency 9.57 % (Fig. 4). The reproducibility of the process and the performances of the devices during samples realization were checked

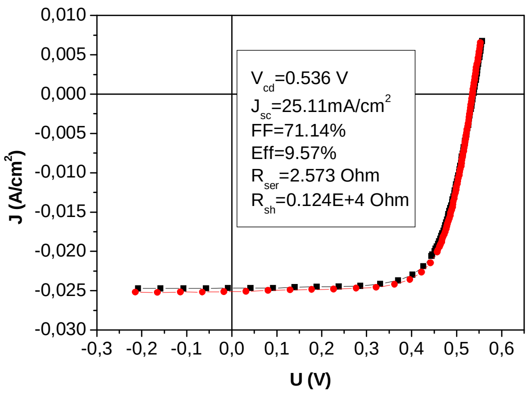

Fig. 4. I-V characteristic of the n^+ITO–SiO_2–nSi cells with active area 8.1cm^2.

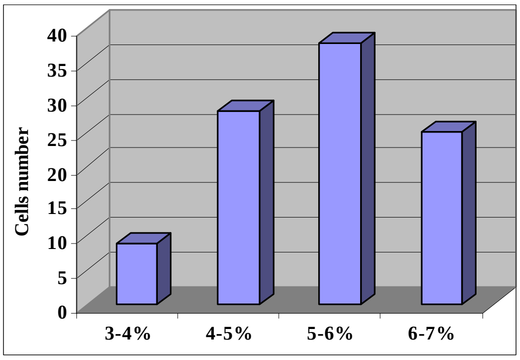

Fig. 5. The efficiency histograms of the ITO-SiO_2-nSi solar cells with 48.6cm^2 active area.

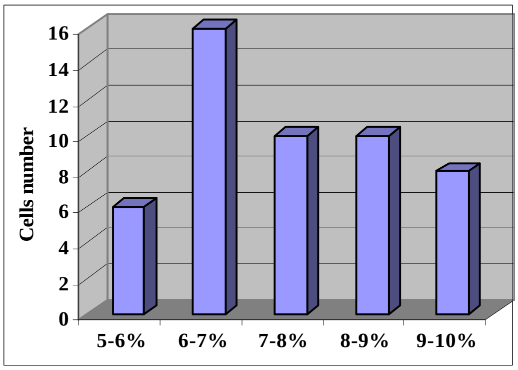

Fig. 6. The efficiency histograms of the ITO-SiO_2-nSi solar cells with 8.1cm^2 active area.

in each batch of samples as well as batch-to-batch. The enlargement of the area of the solar cells up to 48.6 cm^2 leads to the increasing of the series resistance and to the diminishing of the efficiency down to 7 %.

The analysis of the photoelectrical parameters of two sets of SC with different active area permits to obtain the distribution of the fabricated cells according to their efficiency. The respective histograms are presented in Fig. 5 and Fig. 6.

The viability of the developed technology for obtaining solar cells in laboratory conditions was demonstrated by fabrication of the samples of PV modules with the output power up to 30*W*.

III. ITO- pInP Solar cells

Indium phosphide is known to be one of the most preferable materials for the fabrication of solar cells due to its optimum band gap, therefore, it is possible to obtain high efficiency of solar energy conversion into electricity. On the base of InP solar cells have been fabricated with the efficiency of more than 20 % [13]. Moreover, InP based SC are stable under hard radiation conditions. It was shown [14]-[16] that the efficiency of these SC after proton and electron irradiation decreases less than in the case of Si or GaAs based SC. However, due to the high price of InP wafers indium phosphide based SC could not be competitive in terrestrial applications with SC fabricated on other existing semiconductor solar materials such as silicon.

Structures with different crystallographic orientation and holes concentration in the InP substrates have been obtained. The optimum concentration of the charge carriers in pInP substrates is the value 10^{16}cm^{-3}, but the InP wafers with these carrier concentrations and the thickness of 400 μm have a great resistance. For this reason p-p^{+}InP substrates were used in order to obtain efficient solar cells with a low series resistance. In some cases a pInP layer with the thickness up to 4 μm and concentration p = (3...30)·10^{16} cm^{-3} was deposited by the gas epitaxy method from the $In\text{-}PCl_3\text{-}H_2$ system on the (100) oriented surface of InP heavily doped substrate with the concentrations p^{+}= (1...3)·10^{18} cm^{-3} for the fabrication of ITO-pInP-p^{+}InP structures.

The photoelectric properties of these SC have been investigated at the illumination of the heterostructures through the wide gap oxide layer. For all investigated samples the current-voltage characteristics at illumination do not differ from the characteristics of respective homojunction solar cells. The current short-

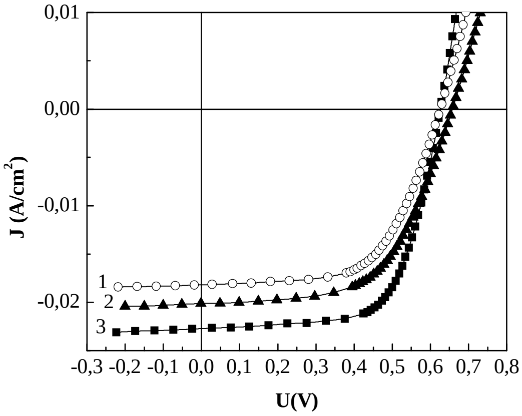

Fig. 7. Load I-V characteristics of Cu/nITO/pInP/Ag:Zn solar cells. 1-before thermal treatment; 2-after thermal treatment in H_2; 3-best parameters after thermal treatment in H_2.

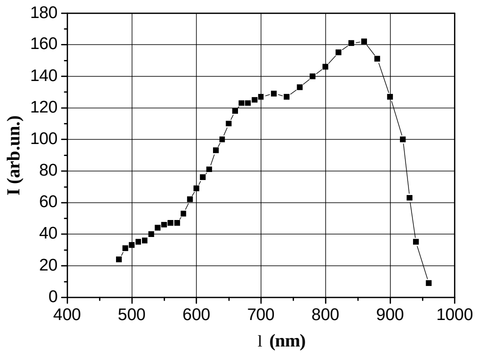

Fig. 8. The spectral photosensitivity curve of Su/n+ITO/pInP/Ag:Zn structure.

circuit I_{sc} linearly depends on the illumination intensity, the open circuit voltage U_{oc} changes with the illumination after the usual logarithmic dependence. The dependence of ITO-InP cells parameters in AM 0 conditions versus InP substrate orientation and hole concentration have been studied. InP wafers with the

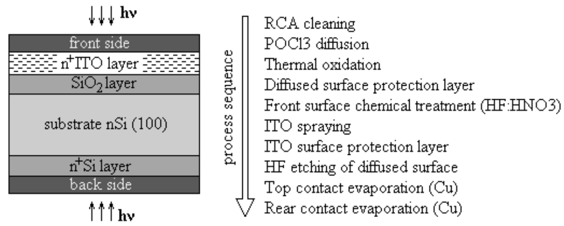

Fig. 9. The schematic view of the bifacial ITO-nSi solar cell and process sequence.

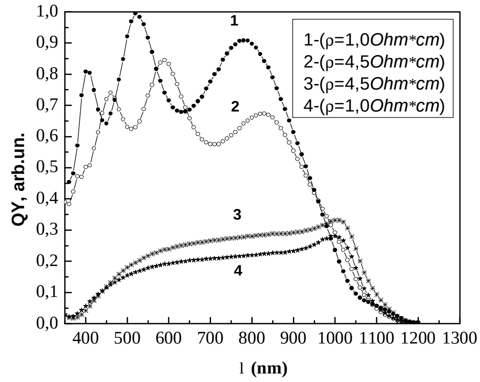

Fig. 10. Spectral distribution of the quantum efficiency: 1, 2-frontal illumination; 3, 4-rear illumination.

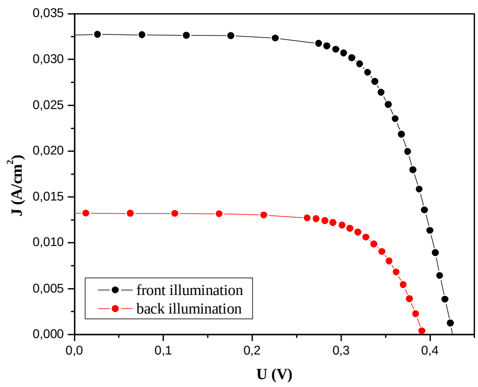

Fig. 11. I-V load characteristics at AM 1.5 illumination.

orientation in (100) and (111) B directions were used for the obtaining of solar cells by the deposition of ITO layers. The respective values of efficiencies are 11.6 % and 10.4 %.

The minimum efficiency has been observed in the case of solar cells obtained by deposition of ITO layers on InP wafers oriented in (111) A direction. To increase the efficiency these solar cells were thermally treated in H_2 atmosphere at the temperature of 350^{0}C during 10 minutes for the diminution of the series resistance [17]. It was shown that before the thermal treatment the following parameters have been obtained under AM 1.5 illumination conditions: U_{oc}= 0.651 V, I_{sc}= 18.12 mA/cm^2, FF = 58 %, Eff.= 6.84 %.(Fig. 7, curve 1). After the thermal treatment the same parameters are U_{oc}= 0.658 V, I_{sc}= 20.13 mA/cm^2, FF = 58 %, Eff.= 7.68 % (Fig. 7, curve 2). The photoelectric parameters of the SC received on InP wafers with concentration $p = 3.10^{17} cm^{-3}$ after the thermal treatment are U_{oc}=0.626 V, I_{sc}= 22.72 mA/cm^2, FF = 71 %, Eff.= 10.1 % (Fig.7, curve 3), that is better than for analogous SC without treatment in H_2. At the same time the thermal treatment in H_2 leads to the undesirable decreasing of the photosensibility in the short wave region of the spectrum. There are necessary supplementary investigations for the clarification of this fact.

The region of the spectral photosensibility of Cu/nITO/pInP/Ag:Zn structure is situated between 470…950 nm (Fig.8). The highest photosensibility is observed at 870nm, which indicates that the maximum contribution in the photosensibility is due to the absorption in InP.

IV. Bifacial ITO-nSi solar cells

Bifacial solar cells (BSC) are promising devices because they are able to convert solar energy coming from both sides of the cell, increasing its efficiency. Different constructions of BSC have been proposed and investigated. In the framework of the classification suggested in [18] the BSC structures could be divided into groups according to the number of junctions: a) two p-n junctions, b) one p-n junction and one high-low junction and, c) just one p-n junction. In all these types of BSC are based on a heteropolar p-n junction. The best results have been obtained in the case of b)-type BSC [19], which were fabricated on the base of n^+-p-p^+ or p^+-n-n^+ structures with back surface field (BSF) at rear contact. In this case it is necessary to obtain two junctions: a heteropolar p-n junction at the frontal side of the silicon wafer and a homopolar n-n^+ or p-p^+ junction at the rear side. Usually these junctions are fabricated by impurity diffusion in the silicon wafer. The diffusion process occurs at temperatures higher then 800^0C and requires special conditions and precise control. In the case of BSF fabrication these difficulties increase since it is necessary to realize the simultaneous diffusion of impurities which have an opposite influence on the silicon properties. Therefore, the problem of protecting the silicon surface from the undesirable impurities appears.

In our case silicon wafers with the electron concentration of 1 Ohm.cm and 4.5 Ohm.cm were used for the fabrication of bifacial SC. A usual back surface field structure consisting of a highly doped n-Si layer obtained by phosphorus diffusion was fabricated on the top of polished side of the wafer by a diffusion process starting from $POCl_3$ gas mixture. The rear n-n^+ junction formation ends with a wet chemical etching of $POCl_3$ residual in a 10 % HF bath. A junction depth of 1 μm has been chosen in order to minimize recombination. To reduce the surface recombination velocity the wafers have been thermally oxidized at a temperature of 850°C. Grids obtained by cuprum (Cu) evaporation in a vacuum were deposited on the frontal and back surfaces for BSC fabrication. The schematic view of the bifacial ITO-nSi solar cell and process sequence is presented in Fig. 9.

The spectral distribution of the quantum efficiency has been studied at frontal and back illumination for samples with different Si resistivity ρ (1.0 Ohm·cm and 4.5 Ohm·cm) (Fig. 10). In the case of frontal illumination, it is seen that in the region of wavelengths from 400 nm to 870 nm the value of QY changes in the limits 0.65–0.95. For back illumination QY is equal to 0.2–0.3 in the same region of the spectrum.

In Fig. 11, the I-V load characteristics at AM 1.5 spectral distribution and 1000 W/m^2 illumination are presented. The photoelectric parameters of the elaborated BSC have been determined in standard AM 1.5 conditions: for the frontal side U_{oc}= 0.425 V, J_{sc}= 32.63 mA/cm^2, FF = 68.29 %, Eff.= 9.47 %, R_{ser}= 2.08 Ohm, R_{sh}= 6.7·10^3 Oh*m*; for the back side U_{oc}= 0.392 V, J_{sc}= 13.23 mA/cm^2, FF = 69.28 %, Eff.= 3.6 %, R_{ser}= 3.40 Ohm, R_{sh}= 1.26·10^4 Ohm.

Conclusions

ITO-nSi and ITO-pInP semiconductor–insulator–semiconductor structures have been produced with a simple spraying technique. The structures obtained in this way may be considered Schottky diodes with a thin insulating layer at the interface. Solar cells on the base of ITO-nSi structures with an active area of 8.1 cm^2 and 48.6 cm^2 have been fabricated and studied. Their quantum efficiency reaches 0.97 at λ = 550 nm. At the AM 1.5 illumination conditions the efficiency is 9.57 % for cells with area of 8.1 cm^2 and 7 % for cells with area 48.6 cm^2. Samples of PV modules with the output power up to 30*W* have been fabricated on the base of ITO-nSi solar cells.

ITO-pInP structures with different crystallographic orientation and whole concentration in the InP substrates have been obtained. The maximum efficiency was obtained in the case of fabrication of ITO-pInP-p^+InP structures using InP wafers oriented in the (100) plane with the hole concentrations p= (3...30)·10^{16} cm^{-3}, p^+= (1...3)·10^{18} cm^{-3}.

A novel type of bifacial cells containing only isotype junctions was elaborated. Their advantages consist of the following: the frontal junction is obtained with a simple spray technique at low temperatures; the double diffusion process is not necessary; the frontal ITO layer is collecting electrode and antireflection coating at the same time. The overall efficiency of the elaborated bifacial solar cells reaches 13 %.

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Transparent Conductive Oxide Layers and Their Application in Solar Energetic

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Сонячні елементи на основі поверхнево-бар'єрних гетероструктур напівпровідник-діелектрик-напівпровідник (SIS) перспективні для перетворення сонячної енергії в електричну через їх низьку собівартість і простоту технології виготовлення. Такі структури отримані осадженням провідного прозорого шару ITO способом піролітичної пульверизації спиртових розчинів хлоридів індію і олова на різні напівпровідникові кристалічні підкладинки. Провідність плівок ITO була $4{,}7\cdot10^{3}$ $Ом^{-1}\cdot см^{-1}$ і пропускання світла ~87%. Як база використовувались пластини nSi і pInP з концентрацією носіїв заряду 10^{15}-10^{17} $см^{-3}$.

Ефективність сонячних елементів ITO-nSi площею 8,1 см^2 в умовах освітлення AM1,5 досягала 9,57%, а у елементів ITO-pInP ≈11%. Виготовлені також двосторонні сонячні елементи ITO-nSi та вивчені їхні спектральні залежності квантової ефективності в діапазоні довжин хвиль 400-870 нм при фронтальному і тильному освітленні; в першому випадку квантова ефективність була в межах 0.60-1, а в другому – 0,20-0,33. Отримані такі фотоелектричні параметри двосторонніх сонячних елементів ITO-nSi: при фронтальному освітленні – U_{oc}= 0.425V, I_{sc}= 32.63 mA/cm^2, FF = 68.29 %, Eff.= 9.47 %, а при тильному – U_{oc}= 0.392 V, I_{sc}= 13.23 mA/cm^2, FF = 69.28 %, Eff.= 3.60 %.

Ключові слова: двосторонній сонячний елемент, ITO, кремній, фосфід індію, TCO.