

EFFECT OF ACTIVE TREATMENTS ON PHOTOVOLTAIC CHARACTERISTICS OF STRUCTURES BASED ON CdTe FILMS

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Photoelectric characteristics of ITO/CdTe structures fabricated by the thermal evaporation in vacuum followed by their deposition in a quasiclosed volume have been studied before and after treatments of various kinds. Some specimens were subjected to a “chloride” treatment, the others were annealed in air. Afterward, the specimens were treated in hydrogen plasma, and they were covered with a thin diamond-like carbon film. The “chloride” treatment of ITO/CdTe structures is shown to result in an increase of the diffusion length of charge carriers in the CdTe layer. The thermal annealing did not affect this parameter, but significantly enhanced the photosensitivity, which means a reduction of the surface recombination rate in the surface CdTe layer. For all considered ITO/CdTe structures obtained by the thermal evaporation in vacuum, the following treatment in hydrogen plasma and the deposition of thin diamond-like films brought about a substantial increase in the diffusion length of charge carriers in the CdTe layer. The ITO/CdTe structures obtained by the thermal vacuum evaporation and treated with hydrogen plasma demonstrated a significant enhancement of their spectral sensitivity in a wavelength range of 400–800 nm, whereas the same effect for structures subjected to the “chloride” treatment was obtained after the sequential hydrogen plasma treatment and the diamond-like carbon film deposition.

1. Introduction

The direct transformation of solar energy into electric one taking advantage of solar cells (SCs) fabricated on the basis of photosensitive semiconductor structures represents an alternative source of energy. However, the cost of electric power produced by the photo-electric method still remains high. Therefore, the problem of its reduction is challenging. A cost reduction of electric energy obtained with the use of semiconductor SCs can be attained in a number of ways, in particular,

by decreasing the cost of semiconductor materials used in the SC fabrication. Therefore, a considerable attention is paid today to researches aimed at improving the design and the production method of thin-film SCs, among which flexible SCs on the basis of CdTe films, which are cheap, seem to be very promising [1–5]. Nowadays, the best specimens of such SCs formed on glass substrates have an efficiency of more than 16.5%.

In order to enhance the efficiency of the photo-electric energy transformation in SCs on the basis of CdTe, various active treatments are used in their fabrication. In particular, the annealing in a chlorine atmosphere invokes a recrystallization in CdTe films with the appearance of grains characterized by a small spread of their dimensions and an increase of the photoluminescence intensity with a peak in a vicinity of 1.4 eV, which corresponds to the width of the forbidden gap in CdTe [6]. The plasma treatment of a CdTe layer by argon ions before the deposition of a rear contact onto it leads to a growth of the short circuit current in obtained SC specimens [7].

However, the influence of various active treatments on the parameters of photosensitive structures created on the basis of CdTe films has not been studied enough yet. Therefore, this work is aimed at comparative researches of the influence of such active treatments as the annealing in air, “chloride” treatment, plasma treatment in hydrogen, and deposition of diamond-like carbon films (DLCFs) on the photo-electric characteristics of structures fabricated on the basis of CdTe films deposited under various conditions.

2. Specimens and Research Technique

In this work, we studied two batches of specimens, which comprised ITO/CdTe structures formed on optical glass K8. The batches differed from each other in the technology applied to the formation of a basic layer, i.e. the CdTe film. Tin and indium oxide (ITO) was deposited onto glass using the method of nonreactive magnetron sputtering. The ITO layer had the thickness $d = 0.5 \mu\text{m}$. It is characterized by a transmittance of 86% in the visible spectral range and a surface resistance of $150 \Omega/\square$. The layer of cadmium telluride was covered with an ITO layer. In the first batch of specimens, the CdTe layer was created using the method of thermal vacuum evaporation; in the second one, it was deposited in a quasiclosed volume.

If the deposition temperature does not exceed 350°C at fabricating the basic stoichiometric CdTe layers by the thermal evaporation in vacuum, transparent polyimide films, which sustain heating up to $T = 450^\circ\text{C}$, can be applied as substrates. Moreover, the method of thermal vacuum evaporation allows a highly productive industrial (“roll”) technology of supplying a flexible substrate to the condensation zone to be implemented.

An advantage of the CdTe film fabrication technology by carrying out the deposition in a quasiclosed volume is a capability of implementing the growth conditions, which are close to the thermodynamically equilibrium ones, which is favored by a small difference between the temperatures of an evaporator and a substrate. The deposition temperature in this technology does not exceed 400°C , which also allows polyimide films to be used as substrates.

The deposition of CdTe films using the thermal evaporation technique was executed on a vacuum installation of the UVN type at an initial vacuum pressure of 10^{-4} Pa. Cadmium telluride films were evaporated from a graphite evaporator 10 cm in length and 1 cm in width, which allowed uniform structures to $10 \times 10 \text{ cm}^2$ in dimensions to be obtained. At the CdTe film deposition, the substrate temperature was $320\text{--}330^\circ\text{C}$, and the evaporator one was $690\text{--}700^\circ\text{C}$. CdTe films created by depositing in a quasiclosed volume were obtained at a deposition temperature of 380°C and an evaporator temperature of 490°C . In both methods, the time of CdTe film deposition was 10 min, and the thickness of the fabricated film was $4\text{--}5 \mu\text{m}$.

After the CdTe layer deposition, the specimens in each of two batches were divided into three groups. The specimens in the first group were not subjected to any treatment, the specimens in the second group were “chloride”-

treated, and the specimens in the third group were annealed in air under the same conditions, as were applied for the second group.

The “chloride” treatment is used in the technology of SC production on the basis of cadmium telluride to improve the crystal and energy structures of the CdTe layer. In this work, CdCl_2 films were deposited onto the surface of CdTe layers of the second-group specimens in both batches using the thermal evaporation technique without heating the substrate; the vacuum installation was used at an initial vacuum level of 2×10^{-3} Pa. Afterward, the obtained multilayered film systems were annealed in air in a closed volume at a temperature of 430°C for 25 min. To remove the reaction products, the annealed specimens were etched in the 5% methanol solution of bromine.

To distinguish between the influence produced by the interaction between the cadmium chloride and cadmium telluride phases and the influence exerted by the thermal annealing in air, we studied a CdTe film, which had been annealed in air in the same regime as at the “chloride” treatment (for 25 min at an annealing temperature of 430°C), which was implemented for the specimens of the third group in both batches. Then, all specimens were treated in hydrogen plasma. Afterward, approximately half an area of the surface of experimental specimens covered with a CdTe layer was protected by a mask, whereas the remaining half was covered with films of amorphous hydrogenated carbon a-C:H:N, using the deposition technique.

The treatment in hydrogen plasma was carried out for 1 min at room temperature, at a power of 175 W in the plasma discharge, and a hydrogen pressure of 25 Pa. DLCFs were deposited using the plasma-enhanced chemical vapor deposition (PE-CVD) method from HF-discharge (13.56 MHz) plasma at a power of 175 W. A gas mixture with the composition $\text{H}_2:\text{N}_2:\text{CH}_4:\text{Ar} = 10:4:3:1$ was used, the gas pressure in the chamber was 100 Pa, and the deposition time was 25 s. Films were deposited at room temperature of the substrate, which was provided by cooling the lower electrode of the reactor, on which the specimens were placed, with water. The deposited DLCFs had a thickness of 0.7 nm.

The spectral dependences of the condenser photo-emf in a wavelength range of $400\text{--}1200 \text{ nm}$ were studied at various stages of experimental specimen fabrication and treatment. They were measured in the regime, which provided an automatic maintaining of a constant level of light illumination. A translucent mica electrode was used at that, which was mechanically pressed to the specimen surface. The spectral dependences were measured

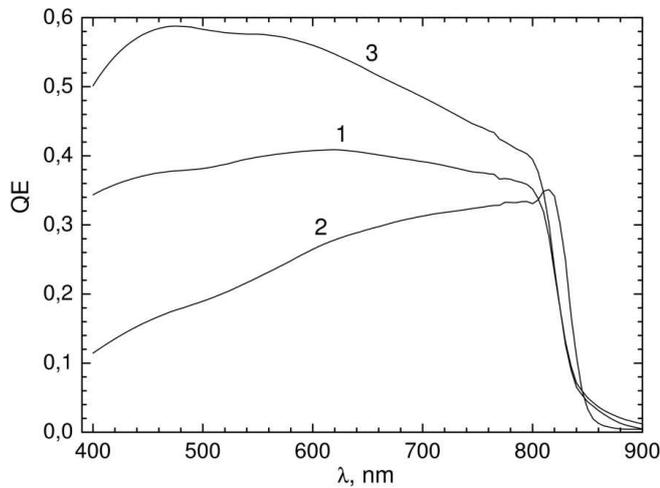


Fig. 1. Spectral dependences of a condenser photo-emf signal normalized to a constant number of light-flux quanta obtained for ITO/CdTe structures (1) without additional treatments, (2) after the “chloride” treatment, and (3) after the annealing in air at $T = 430\text{ }^{\circ}\text{C}$

on an installation for the determination of relative spectral characteristics of photoconverters at the Center for testing photoconverters and photo-electric batteries of V.E. Lashkaryov Institute of Semiconductor Physics of the NAS of Ukraine certified by the State Committee of Ukraine for technical regulation and consumer policy.

3. Experimental Results and Their Discussion

The results of our researches dealing with the spectral dependences of the condenser photo-emf for ITO/CdTe structures formed on glass, when the cadmium telluride layer was deposited in the quasiclosed volume, and obtained before and after the execution of various active treatments are exhibited in Fig. 1. The figure testifies that the spectral dependence obtained for the ITO/CdTe structure after its “chloride” treatment (curve 2) has a sharp decrease at longer waves in comparison with that for a non-treated ITO/CdTe structure (curve 1). This fact points to an increase of the diffusion length of charge carriers in the CdTe layer and comprises a positive result. However, the indicated treatment also gives rise to a substantial reduction of the spectral sensitivity in the short-wave range. This circumstance testifies to an increase of the surface recombination rate on the open surface of a CdTe layer owing to a growth of the concentration of recombination-active centers. The increase of the diffusion length of charge carriers in the CdTe layer after the “chloride” treatment is associated with an

improvement of the crystal and energy structures in it [6].

The spectral dependence obtained for the ITO/CdTe structure after its thermal annealing in air (Fig. 1, curve 3) has a drastic fall in the long-wave spectral region. Here, the spectral dependence for the untreated ITO/CdTe structure is similar, which evidences the absence of a thermal annealing influence on the diffusion length of charge carriers in the CdTe layer. However, in contrast to the results obtained for the ITO/CdTe structure after its “chloride” treatment and if comparing with the untreated ITO/CdTe structure (curve 1), the ITO/CdTe structure subjected to the thermal annealing in air demonstrates a higher sensitivity in the spectral range from 400 nm to the beginning of the drastic fall of its spectral characteristic (820 nm). This fact testifies to a reduction of the surface recombination rate on the open surface of the CdTe layer owing to a decrease of the concentration of recombination-active centers.

The variation in the concentration of recombination-active centers on the open surface of the cadmium telluride layer after the treatments results from a change in the number of dangling bonds at this surface, which is most likely associated with a modification of the stoichiometry in the near-surface region of the CdTe layer. Etching the specimen in a bromine-methanol solution, which is applied after the “chloride” treatment to remove the products of the chemical reaction from the surface, is known to mainly etch cadmium to form a nano-sized layer of tellurium on the surface [8]. In the SC technology, this peculiarity is used to simplify the formation of a tunnel rear contact with the basic cadmium telluride layer [9].

The results of researches dealing with the spectral dependences of the condenser photo-emf for ITO/CdTe structures formed on glass obtained before and after their treatment in hydrogen plasma followed by the deposition of DLCFs are exhibited in Figs. 2 to 7. The results presented in Figs. 2 to 4 were obtained for specimens fabricated using the deposition in a quasiclosed volume, and those in Figs. 5 to 7 for specimens fabricated using the thermal vacuum evaporation method.

The results obtained, which are illustrated in Fig. 2, demonstrate that the spectral sensitivity diminishes after the hydrogen treatment (curve 2) in comparison with the initial one (curve 1) almost in the whole examined interval. The following deposition of a DLCF (curve 3) enhances the spectral sensitivity, but its initial level (curve 1) is not reached. Moreover, after the DLCF having been deposited, the condenser photo-emf signal

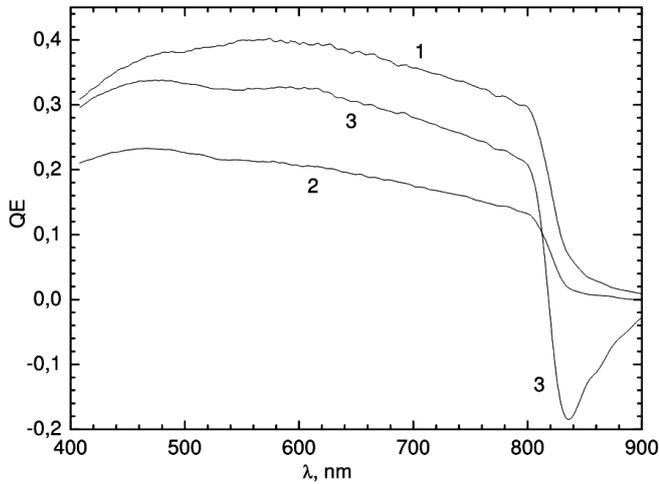


Fig. 2. Spectral dependences of a condenser photo-emf signal normalized to a constant number of light-flux quanta obtained for the ITO/CdTe structures formed under quasiclosed-volume conditions: (1) the initial spectral dependence, (2) after the hydrogen treatment, (3) after the hydrogen treatment and the DLCL deposition

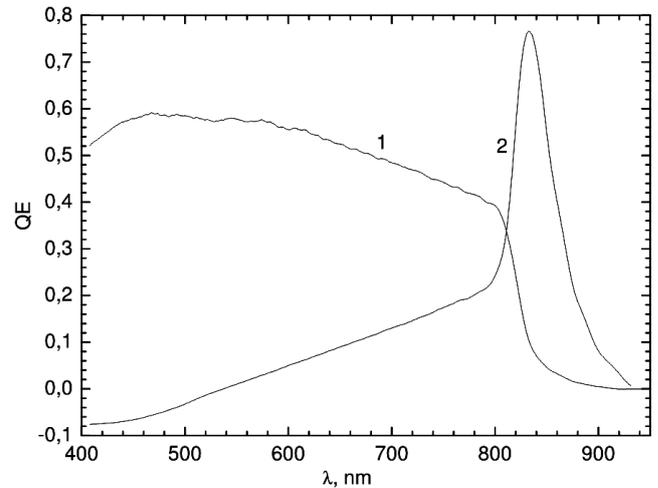


Fig. 4. Spectral dependences of a condenser photo-emf signal normalized to a constant number of light-flux quanta obtained for the ITO/CdTe structure preliminarily annealed in air at $T = 430$ °C: (1) the initial spectral dependence, (2) after the hydrogen treatment and the DLCL deposition

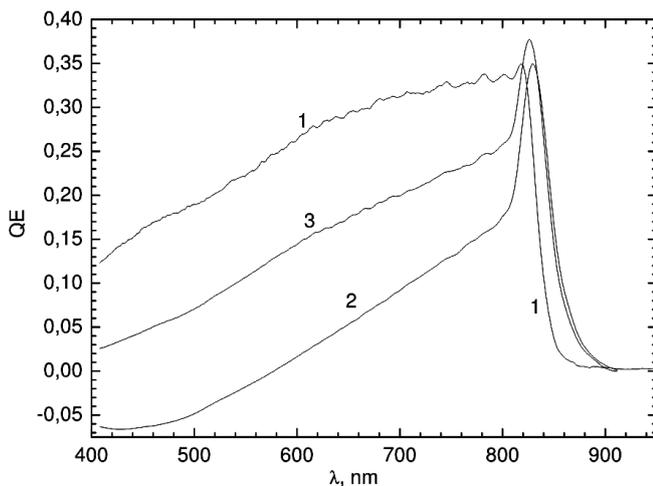


Fig. 3. The same as in Fig. 2, but for the ITO/CdTe structure "chloride"-treated previously

changes its sign to the negative one in the long-wave spectral section.

In Fig. 3, the spectral dependences of the condenser photo-emf are depicted, which were obtained for the ITO/CdTe structure preliminarily "chloride"-treated, as was described above. From Fig. 3, one can see that, in this case, the hydrogen treatment also brings about a reduction of the spectral sensitivity practically in the whole studied range. The following deposition of a DLCL increases the spectral sensitivity, but its initial level (curve 1) is not attained, as was in the previous

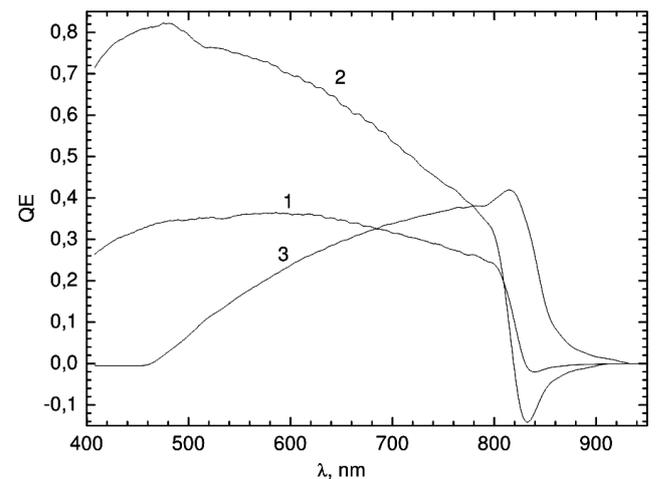


Fig. 5. Spectral dependences of a condenser photo-emf signal normalized to a constant number of light-flux quanta obtained for the ITO/CdTe structure formed by the thermal vacuum evaporation: (1) the initial spectral dependence, (2) after the hydrogen treatment, (3) after the hydrogen treatment and the DLCL deposition

case (Fig. 2). In addition, the condenser photo-emf signal changes its sign to the negative one in the short-wave spectral region after the hydrogen treatment. From Fig. 3, one can see that the spectral dependence obtained for the ITO/CdTe structure after its hydrogen treatment (curve 2) has a drastic fall in the long-wave spectral region in comparison with the initial one (curve 1), with this result remaining unchanged after the DLCL deposition as well.

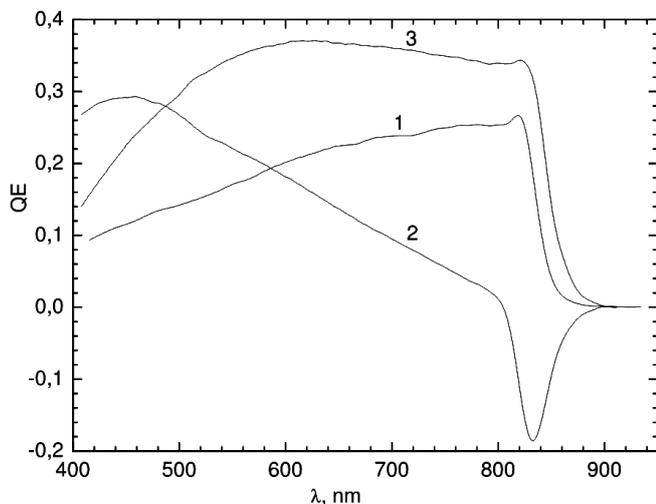


Fig. 6. The same as in Fig. 5, but for the preliminarily "chloride"-treated ITO/CdTe structure

Figure 4 demonstrates the spectral dependences of the condenser photo-emf obtained for the ITO/CdTe structure, which was preliminarily subjected to the thermal annealing in air, which was described above. In this case, the hydrogen treatment and the subsequent DLCF deposition give rise to the following variations in the spectral dependence: a considerable reduction of the spectral sensitivity in the wavelength range below 800 nm, a shift of the drastic fall in the spectral characteristics toward long waves, and the sign inversion for the condenser photo-emf signal to the negative one.

In Fig. 5, the spectral dependences of the condenser photo-emf obtained for the ITO/CdTe structure fabricated by the method of thermal evaporation in vacuum before and after its hydrogen treatment followed by the deposition of a DLCF layer are depicted. The results presented in this figure make it evident that the hydrogen treatment leads to a substantial growth in the spectral sensitivity practically in the whole examined range. The following deposition of a DLCF reduces the spectral sensitivity and gives rise to a shift of the long-wave fall in the spectral dependence toward longer-wave region. In addition, the condenser photo-emf signal changes its sign to the negative one in the long-wave spectral interval.

Figure 6 illustrates the spectral dependences of the condenser photo-emf obtained for the ITO/CdTe structure, which was preliminarily "chloride"-treated, as was described above, and then subjected to the thermal annealing. One can see that the hydrogen treatment enhances the short-wave spectral sensitivity, but the condenser photo-emf signal changes its sign to the negative one in the long-wave spectral region. The following de-

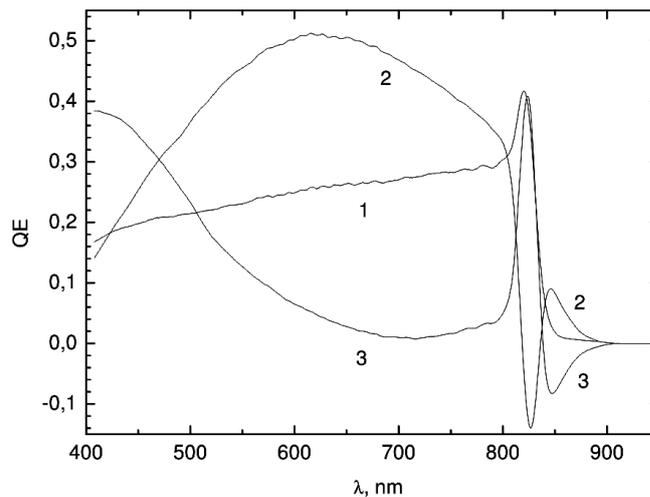


Fig. 7. The same as in Fig. 5, but for the ITO/CdTe structure preliminarily annealed in air at $T = 430$ °C

position of a DLCF changes the situation considerably. In comparison with the spectral dependence obtained before the treatments (curve 1), the spectral dependence obtained after the hydrogen treatment and the DLCF deposition (curve 3) is characterized by a higher photosensitivity in the whole spectral range under consideration, no variation in the condenser photo-emf sign, and a shift of the drastic fall in the spectral characteristic toward longer wavelengths.

In Fig. 7, the spectral dependences of the condenser photo-emf obtained for the ITO/CdTe structure, which was preliminarily subjected to the thermal annealing in air at $T = 430$ °C, are shown. It is evident that, in this case, the hydrogen treatment leads to an increase of the spectral sensitivity, but a considerable fall of the sensitivity in the short-wave region also takes place. In addition, after the treatments, the condenser photo-emf signal changes its sign to the negative one in the long-wave spectral region.

The analysis of the results obtained, which are presented in Figs. 2 to 7 allowed us to establish the following features. The inversion of the condenser photo-emf signal sign to the negative one, which was observed in some cases, testifies that there arises a barrier in the studied structures subjected to certain active treatments or their combinations. After manufacturing SCs on the basis of the structures concerned, this barrier will be electrically connected oppositely to the working $p - n$ -transition and, consequently, will negatively affect the photocurrent magnitude by reducing it down, as well as the efficiency of those SCs. The change of the condenser photo-emf signal sign, which took place in both

short- and long-wave parts of the spectral characteristic, means that this barrier can arise near both the open surface of the CdTe layer and the surface protected by the ITO layer. The other changes in the spectral sensitivity of studied specimens under the influence of active treatments stem from a variation of the concentration of recombination-active centers at the open surface of the CdTe layer, which gives rise to a variation of the surface recombination rate, and with a reduction of the recombination losses in bulk, which leads to an increase in the diffusion length of charge carriers in the CdTe layer (a shift of the drastic fall in some spectral dependences toward the longer-wave spectral region testifies to that). The changes in the concentration of recombination-active centers at the open surface of the CdTe layer are associated with variations in the number of dangling bonds at it owing to a modification of the stoichiometry in the near-surface region of the cadmium telluride layer, whereas a reduction of the bulk recombination losses after certain active treatments is stimulated by improvements in the crystal and energy structures of the CdTe layer.

A comparison of experimental results of our researches concerning the influence of active treatments on the spectral dependences of the condenser photo-emf in ITO/CdTe structures formed under quasiclosed volume conditions showed that the hydrogen treatment of specimens, which were not preliminarily subjected to any treatment or were “chloride”-treated, reduced the spectral sensitivity practically in the whole studied range of wavelengths; the sensitivity can be raised a little by depositing a DLCF, although not to the initial level that was observed before the plasma treatment of specimens (see Figs. 2 and 3). At the same time, the hydrogen treatment of and the following DLCF deposition on ITO/CdTe structures previously subjected to the “chloride” treatment or the thermal annealing in air (Figs. 3 and 4) increased the diffusion length of charge carriers in the CdTe layer.

The experimental results obtained for ITO/CdTe structures formed by the method of thermal vacuum evaporation showed that the hydrogen treatment of specimens, which were not previously subjected to any treatment or were thermally annealed in air (Figs. 5 and 7), enhanced the spectral sensitivity practically in the whole studied range of wavelengths, and the following deposition of a DLCF decreased it. In addition, if the ITO/CdTe structure were not preliminarily treated (Fig. 5), the hydrogen treatment and the deposition of DLCF led to a growth of the diffusion length of charge carriers in the CdTe layer. For the previously “chloride”-

treated ITO/CdTe structure (Fig. 6), its hydrogen treatment followed by the DLCF deposition resulted in a substantial increase of the spectral sensitivity in the whole studied range of wavelengths.

Different results obtained for the experimental ITO/CdTe specimens formed by applying either the deposition in a quasiclosed volume or the method of thermal vacuum evaporation and sequentially subjected to the hydrogen treatment and the DLCF deposition originate from the structural differences between cadmium telluride layers obtained under different conditions. The results of our previous researches testify that the layers fabricated by the deposition in a quasiclosed volume were characterized by a considerably higher quality of its crystal structure (larger grain dimensions, a lower concentration of crystal structure defects, and lower micro- and macrostresses) both before and after the “chloride” treatment in comparison with the basic layers obtained by the thermal evaporation in vacuum [10–12]. Therefore, our results testify that the recombination parameters for basic cadmium telluride layers with different qualities of their crystal structures can be improved, provided that the hydrogen treatment of ITO/CdTe heterosystems and the following DLCF deposition are executed in optimized regimes.

4. Conclusions

To summarize, the “chloride” treatment of the ITO/CdTe structure was found to increase the diffusion length of charge carriers in the CdTe layer. The application of only the thermal annealing in air at a temperature of 430 °C does not affect this parameter, but substantially enhances the photosensitivity in the spectral range from 400 nm to the beginning of its drastic fall (820 nm), which testifies to a reduction of the surface recombination rate at the CdTe layer surface. Some combinations of such active treatment procedures as the thermal annealing in air, the “chloride” treatment, the treatment in hydrogen plasma, and the deposition of thin diamond-like carbon films were demonstrated to increase the diffusion length of charge carriers in the cadmium telluride layer of the ITO/CdTe structures fabricated by both the methods of thermal vacuum evaporation and the deposition in a quasiclosed volume. For the ITO/CdTe structures, which were obtained by the thermal vacuum evaporation and either were not subjected to any additional treatment or were thermally annealed in air, the treatment in hydrogen plasma was found to considerably increase their spectral sensitivity in a wavelength range of 400–800 nm. At the same time, for the “chloride”-

treated structures, a substantial growth of their spectral sensitivity in this range is reached after the sequential treatment in hydrogen plasma and the deposition of diamond-like films.

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ВПЛИВ АКТИВНИХ ОБРОБОК НА ФОТОЕЛЕКТРИЧНІ ХАРАКТЕРИСТИКИ СТРУКТУР НА ОСНОВІ ПЛІВОК CdTe

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Резюме

Досліджено фотоелектричні характеристики структур ІТО/CdTe, виготовлених методом термічного вакуумного випаровування та шляхом осадження у квазізамкненому об'ємі до та після різних обробок. Частина зразків проходила "хлоридну" обробку, інша – відпал на повітрі. Після цього проводилась обробка зразків у плазмі водню та нанесення на них тонкої алмазоподібної плівки. Показано, що проведення "хлоридної" обробки структур ІТО/CdTe приводить до збільшення дифузійної довжини носіїв заряду у шарі CdTe. Проведення термовідпалу не впливає на значення дифузійної довжини носіїв заряду у шарі CdTe, але значно підвищує фоточутливість, що свідчить про зменшення на поверхні шару CdTe швидкості поверхневої рекомбінації. Шляхом комбінації термовідпалу, "хлоридної" обробки, плазмової обробки у водні та нанесення тонких алмазоподібних плівок отримано збільшення довжини дифузії носіїв заряду у шарі CdTe в усіх досліджуваних структурах ІТО/CdTe. На структурах ІТО/CdTe, отриманих термічним вакуумним випаровуванням, обробка у плазмі водню приводила до значного збільшення спектральної чутливості у діапазоні довжин хвиль 400–800 нм, а на структурах, які пройшли "хлоридну" обробку, значне збільшення спектральної чутливості досягалось після обробки у плазмі водню та нанесення алмазоподібних плівок.