Effect of Nanosized Tin Oxide Layer on the Efficiency of Photovoltaic Processes in Film Solar Cells Based on Cadmium Telluride

G.S. Khrypunov¹, O.V. Pirohov¹, D.A. Kudiy¹, R.V. Zaitsev¹, A.L. Khrypunova¹, V.A. Gevorkyan², P.P. Gladyshev³

¹ National Technical University "Kharkiv Polytechnic Institute", 21, Frunze Str., 21, 61002 Kharkiv, Ukraine
 ² Russian-Armenian (Slavonic) University, 123, Hovsep Emin Str., 0051 Yerevan, Armenia
 ³ "Dubna International University", 19, Universitetskay Str., 141980 Dubna, Moscow Region, Russia

(Received 02 October 2014; published online 25 March 2015)

The influence of the thickness of the nanosized layer on the efficiency of photoelectric processes in solar cells (SC) ITO / SnO_2 / CdS / CdTe / Cu / Au formed on different substrates was investigated. For device structures formed on the glass substrates, the maximum efficiency of 11.4 % is achieved when thickness of the tin oxide layer is 80 nm. For flexible solar cells formed on a polyimide film, the maximum efficiency of 10.8 % is observed when thickness of the tin oxide layer is 50 nm. This paper discusses the physical mechanisms of the observed differences in efficiency.

Keywords: Thin film solar cells, Glass and flexible substrates, Tin dioxide, Cadmium sulfide and cadmium telluride.

PACS numbers: 81.40.Ef, 61.05. - Cp, 68.57. - Hk

1. INTRODUCTION

Formation of flexible device structures by the replacement of the glass substrate by the transparent thermostable polyimide film is a modern innovation direction of the improvement of promising for large-scale application film solar cells (SC) based on cadmium sulfide and telluride [1]. Such flexible device structures have a record power per unit weight and can be mounted on the surfaces of any forms [2] that allows to increase the SC market segment on account of the application in space and defense technologies. Radiation resistance of cadmium telluride allows to use these SC even for electric power supply of gamma detector [3]. Recently, decrease in the cadmium sulfide layer thickness for the increase in the density of photon flow coming to the base cadmium telluride layer is one of the approaches to increase the efficiency of CdS / CdTe based SC formed on the glass substrate [4]. At that, dielectric nanosized interlayers of undoped oxides of different metals are deposited on the frontal electrode surface to prevent shunting of device structure as a result of the electrical contact of the base layer and frontal electrode through discontinuities in the cadmium sulfide layer during the SC formation [5]. For the realization of this approach to the improvement of the structural and technological solutions of flexible SC based on CdS / CdTe, we have carried out in the given work a comparative study of the influence of nanosized interlayers of tin oxide on the efficiency of photoelectric processes of the mentioned device structures formed on the glass and flexible substrates.

2. DESCRIPTION OF THE OBJECTS AND INVESTIGATION METHODS

Film SC based on CdS / CdTe were formed on the glass substrates (Nippon sheet Company) and flexible polyimide films (Upilex Company) with industrially deposited films of indium and tin oxides (ITO). ITO film thickness was equal to 120 nm. Then, thin films of undoped tin oxide of different thickness were deposited in

the vacuum plant VUP-5M on the ITO layers by the nonreactive magnetron sputtering method on direct current at the substrate temperature of 300 °C. Cadmium sulfide and telluride films were deposited by the thermal vacuum evaporation method without vacuum failure at the substrate temperature of 200 °C and 300 °C, respectively, after formation of the frontal electrodes in the upgraded vacuum device UVN. In this case, cadmium sulfide thickness (dcds) was equal to 0.2 µm, cadmium telluride one $-4 \,\mu m$. Then, by the technique described in [6] "chloride" treatment was performed. After "chloride" treatment, etching of the base layer was carried out in the bromine solution and in methanol, and then, backside film Cu / Au contacts were formed by the thermal vacuum evaporation method. Determination of the output parameters and light diode characteristics of SC was realized by the approximation of the experimental light CVC by the theoretical expression [7]. During the analytical processing, roof-mean-square deviation of the theoretical CVC from the experimental one did not exceed 10^{-8} that corresponds to the relative error in the determination of the output parameters and light diode characteristics on the level not more than 1 %.

Investigation of the spectral dependence of the quantum efficiency coefficient $Q(\lambda)$ allows to analyze the integral efficiency of the photoelectric processes, namely, generation, diffusion, drift, separation and collection of the generated under the action of light non-equilibrium charge carriers subject to the incident radiation energy. Therefore, along with the measurement of the light CVC these investigations are necessary for the optimization of the structural and technological SC solutions. Quantum efficiency coefficient Q is determined as the ratio of the number of charge carriers contributing into photocurrent to the general number of photons coming at the active SC surface [8]. There is a functional connection between the short-circuit photocurrent I_{sc} and the value of $Q(\lambda)$ described at a sufficiently large shunting resistance $R_{\rm sh}$ by the relation [7]

$$I_{\rm sc} = e \int Q(\lambda) \cdot N(\lambda) d\lambda - I_{\rm d}, \tag{1}$$

G.S. KHRYPUNOV, O.V. PIROHOV, D.A. KUDIY, ET AL.

where λ is the light wavelength; λ_k photoelectric threshold; $N(\lambda)$ is the rate of photon arrival on the SC surface; I_d is the SC diode current.

In the real conditions, intensity of solar radiation coming on the SC surface is such that $I_d \ll I_{sc}$ for the observed value of the device structure series resistance, therefore

$$I_{\rm sc} = eQ(\lambda) \cdot N(\lambda). \tag{2}$$

The value of $N(\lambda)$ can be expressed through the light intensity $I_1(\lambda)$ coming on the SC surface:

$$Q(\lambda) = [I_{\rm sc}(\lambda) \cdot E(\lambda)] / [eS \cdot I_{\rm l}(\lambda)].$$
(3)

The values of $I_{\rm sc}(\lambda)$ in relation (3) were determined experimentally; $I_{\rm l}(\lambda)$ is the characteristic of the 500 W incandescent lamp which was used as the light source. In the investigations of the spectral dependence of the photoresponse, SC was located at the output slit of the double monochromator, and measurement of the shortcircuit current with a gradual change of the incident radiation wavelength was performed.

3. DESCRIPTION AND ANALYSIS OF THE RESULTS

Light CVC of ITO / SnO2 / CdS / CdTe / Cu / Au SC formed on the glass substrates and flexible polyimide films have been investigated. The following output SC parameters were calculated by the light CVC: short-circuit current density (J_{sc}) , open-circuit voltage (U_{oc}) , fill factor of the light CVC (FF) and efficiency (η). Then, by the analytical processing of the light CVC we have determined the light diode characteristics, namely, photocurrent density (J_{ph}) , diode saturation current density (J_0) , diode ideality coefficient (A), series (R_s) and shunting $(R_{\rm sh})$ resistances calculated per unit of the device structure active area [9]. Study of the output parameters and light diode characteristics of SC allowed to separate the intervals of the tin oxide layer thicknesses corresponding to the change of the physical mechanisms of influence of the dielectric interlayer on the efficiency of the device structure photoelectric processes. The typical light CVC of ITO / SnO₂ / CdS / CdTe / Cu / Au SC are shown in Fig. 1a, b.

The output parameters and light diode characteristics of ITO / SnO_2 / CdS / CdTe / Cu / Au SC formed on the glass substrates and polyimide films are represented in Table 1.

As the analysis of Table 1 shows, for cadmium sulfide layer thickness of 0.2 µm, growth of the tin oxide layer thickness ($d_{\rm Sn02}$) to 80 nm leads to the increase in the efficiency coefficient of ITO / SnO₂ / CdS / CdTe / Cu / Au SC formed on the glass substrates to $\eta = 11.4$ %. Here, open-circuit voltage increases to $U_{\rm oc} = 765$ mV, fill factor of the light CVC increases to FF = 0.71 and shortcircuit current density decreases to $J_{\rm sc} = 20.9$ mA/cm². According to the results of the mathematical modeling of the quantitative impact of the light diode characteristics on the SC efficiency, whose procedure is described in the work [10], growth of η is conditioned by the increase in the shunting resistance to $R_{\rm sh} = 800$ Ohm cm² and decrease in the diode saturation current density to $J_0 = 2.5 \cdot 10^{-8}$ A/cm². Further increase in the layer thickness leads to the decrease in the SC efficiency. The results of the modeling show that this is conditioned by the growth of the series resistance and decrease in the photocurrent density.



а



b

Fig 1 – Light CVC of the ITO / SnO₂ / CdS / CdTe / Cu / Au SC. a – glass substrates: $1 - d_{\text{SnO2}} = 50 \text{ nm}$, $2 - d_{\text{SnO2}} = 80 \text{ nm}$ and $3 - d_{\text{SnO2}} = 120 \text{ nm}$ and b – flexible substrates: $1 - d_{\text{SnO2}} = 0 \text{ nm}$, $2 - d_{\text{SnO2}} = 50 \text{ nm}$ and $3 - d_{\text{SnO2}} = 80 \text{ nm}$

It was established that in ITO / SnO₂ / CdS / CdTe / Cu / A SC formed on the polyimide films growth of the tin oxide layer thickness to $d_{\text{SnO2}} = 50 \text{ nm}$ leads to the increase in the efficiency coefficient to $\eta = 10.8$ %. Here, open-circuit voltage increases to $U_{oc} = 792 \text{ mV}$, fill factor of the light CVC increases to FF = 0.70 and shortcircuit current density decreases to $J_{sc} = 19.5 \text{ mA/cm}^2$. According to the results of the mathematical modeling of the quantitative impact of the light diode characteristics on the SC efficiency, growth of η is conditioned by the increase in the shunting resistance to the value of $R_{\rm sh} = 1100 \text{ Ohm cm}^2$ and decrease in the diode saturation current density to $J_0 = 1.5 \cdot 10^{-8} \text{ A/cm}^2$. Further increase in the layer thickness leads to the decrease in the SC efficiency. The results of the modeling show that positive influence on the SC efficiency on account of the continuing increase in the shunting resistance and decrease in the diode saturation current density ceases to compensate the negative influence on the SC efficiency due to the increase in the series resistance and decrease in the photocurrent density.

Output parameters	$d_{\rm CdS} = 0.2 \ \mu m \ (1)$			$d_{\rm CdS} = 0.2 \ \mu m \ (2)$		
and light diode characteristics	$d_{ m SnO2}$ = 50 nm	$d_{ m SnO2}$ = 80 nm	$d_{ m SnO2}$ = 120 nm	$d_{ m SnO2}$ = 0 nm	$d_{ m SnO2}$ = 50 nm	$d_{ m SnO2}$ = 80 nm
$J_{ m sc},{ m mA/cm^2}$	21.4	20.9	20.5	19.8	19.5	19.0
$U_{ m oc},{ m mV}$	649	765	760	735	792	802
FF, rel. units	0.52	0.71	0.63	0.52	0.70	0.60
$\eta, \%$	7.2	11.4	9.9	7.4	10.8	9.2
$J_{ m ph}$, mA/cm 2	21.6	21.3	20.7	20.2	19.6	19.2
$R_{ m s}, m Ohmcm^2$	1.8	2.8	4.2	1.5	2.1	2.3
$R_{ m sh}, m Ohmcm^2$	450	800	700	600	1100	1300
A, rel. units	2.7	2.1	2.2	2.5	1.9	1.8
J_0 , A/cm ²	$4.3 \cdot 10^{-7}$	$2.5 \cdot 10^{-8}$	$2.8 \cdot 10^{-8}$	$6.7 \cdot 10^{-8}$	$1.5 \cdot 10^{-8}$	$9.3 \cdot 10^{-9}$

Table 1 – Output parameters and light diode characteristics of ITO / SnO_2 / CdS / CdTe / Cu / Au SC formed on the glass (1) and polyimide (2) substrates

The performed investigations of the output parameters and light diode characteristics were supplemented by the study of the spectral dependences of the quantum efficiency coefficients of the typical SC (Fig. 2, 3). It was experimentally established that photosensitivity range of ITO / SnO₂ / CdS / CdTe / Cu / Au SC formed on the glass substrates is equal to 300-900 nm (Fig. 2a). Photosensitivity growth in the spectral range of 300-500 nm and photosensitivity decrease in the spectral range of 500-900 nm is observed with increasing tin dioxide layer thickness to 80 nm (Fig. 2b). Since light absorption depth decreases with decreasing wavelength, then increase in the photoresponse contribution in the spectral range of 300-400 nm implies the decrease in the surface recombination with increasing tin oxide layer thickness to 80 nm. This is also confirmed by the observed growth of the shunting resistance and decrease in the diode saturation current density. Further increase in the tin oxide layer thickness leads to the decrease in the photosensitivity in the whole spectral range of 300-800 nm (see Fig. 2c).

In contrast to the device structures on the glass substrates, photosensitivity spectral interval of ITO / SnO₂ / CdS / CdTe / Cu / Au SC formed on the polyimide films is equal to 400-900 nm (Fig. 3). Here, reduction in the photosensitivity in the whole spectral range is observed with increasing tin oxide layer thickness. Optical study of the transmission spectra of the glass substrate, polyimide film and cadmium sulfide (Fig. 4) indicates that shift of the photosensitivity short-wavelength limit of flexible SC towards the long-wave region is conditioned by the absence of transmission of polyimide film in the spectral range of 300-400 nm in spite of the fact that at the cadmium sulfide layer thickness of 0.2 µm a considerable part of photons with the energy larger than the CdS band gap comes to the cadmium telluride base layer. Lower transmission coefficient of polyimide film in the spectral range of 400-900 nm leads to the experimentally observed decrease in the short-circuit current density of flexible SC. At the same time, flexible SC have larger values of the shunting resistance and smaller diode saturation current densities that, from our point of view, is connected with the transition from the glass substrate to the polyimide film. Application of sodium-containing glasses as the substrates induces diffusion of sodium to the device structure [10]. According to the literature [11], in the base CdTe layers a sodium atom, which occupies a position of cadmium (Na_{Cd}), represents an acceptor,



Fig. 2 – Spectral dependences of the quantum efficiency coefficient: a) $Q(\lambda)$ of ITO / SnO₂ / CdS / CdTe / Cu / Au SC formed on the glass substrates: $\bullet - d_{\text{SnO2}} = 50 \text{ nm}$, $\blacksquare - d_{\text{SnO2}} = 80 \text{ nm}$, $\blacktriangledown - d_{\text{SnO2}} = 120 \text{ nm}$; b) $Q(\lambda)d_{\text{SnO2}} = 80 \text{ nm}$, $Q(\lambda)d_{\text{SnO2}} = 50 \text{ nm}$ and c) $Q(\lambda)d_{\text{SnO2}} = 80 \text{ nm}$, $Q(\lambda)d_{\text{SnO2}} = 120 \text{ nm}$

and Nac_d - Cl_{Te} donor-acceptor pair is a deep recombination center. Appearance of these structural defects is predicted by the diffusion of Na from the glass sodium-containing substrates to the base layer.

Presence of Na_{Cd} and Na_{Cd}-Cl_{Te} on the grain-boundary surface in the region of the heterojunction shunts the separating barrier that decreases $R_{\rm sh}$. Formation in the volume of grains of the base layer of defect complexes Na_{Cd}-Cl_{Te} decreases the lifetime of the non-equilibrium charge carriers that leads to the J_0 increase. Layer-bylayer elemental analysis of ITO / CdS / CdTe / Cu / Au heterosystems (see Fig. 5a) formed on the glass substrates implies a considerable concentration of Na in the base CdTe layers. Layer-by-layer elemental analysis of ITO / SnO₂ / CdS / CdTe / Cu / Au heterosystems (Fig. 5b) shows that use in the SC design of the tin oxide interlayer leads to the substantial decrease in the sodium concentration in the cadmium telluride layer.



Fig. 3 – Spectral dependences of the quantum efficiency coefficient: a) $Q(\lambda)$ of ITO /SnO₂ / CdS / CdTe / Cu / Au SC formed on the flexible substrates: $\bullet - d_{\text{SnO2}} = 0 \text{ nm}, \blacksquare - d_{\text{SnO2}} = 50 \text{ nm}, \blacksquare - d_{\text{SnO2}} = 80 \text{ nm};$ b) $Q(\lambda)d_{\text{SnO2}} = 50 \text{ nm}; Q(\lambda)d_{\text{SnO2}} = 0 \text{ nm}$ and c) $Q(\lambda)d_{\text{SnO2}} = 50 \text{ nm}; Q(\lambda)d_{\text{SnO2}} = 80 \text{ nm}$



Fig. 4 – Spectral dependences of the transmission coefficient: 1 - glass substrate, 2 - polyimide film, 3 - CdS film



Fig. 5 – Distribution of sodium in SC formed on the glass substrates: a) ITO / CdS / CdTe / Cu / Au, b) ITO / SnO₂ / CdS / CdTe / Cu / Au at $d_{SnO2} = 50$ nm

4. CONCLUSIONS

It was established that the substrate type defines the optimal thickness of the tin oxide dielectric interlayer at which the maximum efficiency coefficient is observed in ITO / SnO₂ / CdS / CdTe / Cu / Au SC. For the device structures formed on the glass substrates the maximum efficiency of 11.4 % is achieved for the tin oxide layer thickness of 80 nm. For flexible SC formed on the polyimide films the maximum efficiency of 10.8 % is observed for the tin oxide layer thickness of 50 nm.

EFFECT OF NANOSIZED TIN OXIDE LAYER ON THE EFFICIENCY ...

It is experimentally shown that in the device structures formed on the glass substrates one observes the diffusion of Na to the base CdTe layer that leads to the experimentally fixed increase in the diode saturation current density and decrease in the quantum efficiency coefficient in the short-wave spectral region of photosensitivity. Therefore, large optimal values of the dielectric interlayer for SC on the glass substrates are conditioned by the fact that in such device structures nanosized tin dioxide layers not only prevent shunting of the heterojunction due to the contact of cadmium telluride

REFERENCES

- W.L. Rance, J.M. Burst, D.M. Meysing, C.A. Wolden, M.O. Reese, L.A. Gessert, W.K. Metzger, P. Cimo, T.M. Barnes, *Appl. Phys. Lett.* **104**, 143903 (2014).
- J. Perrenoud, S. Buechleler, A.N Tiwari, Proceedings 34th IEEE Photovoltaic Specialist Conference Materials Research Society Spring Meeting, 695 (Philadelphia USA: 2013).
- A.I. Skrypnyk, A.A. Zakharchenko, M.A. Khazhmuradov, E.M. Prokhorenko, V.F. Klepikov, V.V. Lytvynenko, Problems of Atomic Science and Technology. Series: Nuclear Physics Investigation No3(85), 231 (2013).
- 4. H.A. Mohamed, J. Appl. Phys. 113, 093105 (2013).
- 5. A. Fuchs, H.J. Schimper, A. Klein, W. Jaegermann, Energ.

and frontal ITO electrode, but also serve the diffusion barrier for sodium.

Decrease in the ITO / SnO2 / CdS / CdTe / Cu / Au SC efficiency coefficient during the glass/flexible substrate transition is conditioned by the absence of transmission of polyimide film in the spectral range of 300-400 nm and lower transmission coefficient in the spectral range of 400-900 nm that leads to the corresponding decrease in the quantum efficiency coefficient in the whole range of photosensitivity and decrease in the short-circuit current density.

Procedia **10**, 149 (2011).

- 6. G.S. Khripunov, Semiconductors 40, 113 (2006).
- 7. H.S. Raushenbach, *Solar Cells Array Design* (New York: Litton Education Publishing: 1980).
- D.L. Batzner, G. Agostinelli, N. Campo, A. Romeo, H. Zogg, A.N. Tiwari, *Thin Solid Films* 431-432, 421 (2003).
- G.S. Khripunov, A.V. Mepiyts, Ukr. J. Phys. 49 №12, 1188 (2004).
- L. Kranz, J. Perrenoud, F. Pianezzi, C. Gretener, P. Rossbach, S. Buechleler, A.N. Tiwari, *Sol. Energ. Mat. Sol. C.* 105, 213 (2012).
- 11. S.Y. Wei, S.B. Znang, phys. status solidi b 229, 305 (2002).