

Шляхом експериментальних досліджень вивчені механізми деградації CE CdTe в процесі експлуатації. Визначено два механізми деградації таких сонячних елементів. Перший обумовлений генерацією дефектів в області переходу, яка обумовлена надлишковими носіями заряду і дефектами. Другий – зростанням величини тильного бар'єру. Дослідження вольт-амперних і вольт-фарадних характеристик сонячних елементів дозволили запропонувати модель деградації сонячних елементів на основі CdTe. Встановлено, що наявність міді у складі тильного контакту пов'язано з найкращою початковою ефективністю однак і найбільш швидкою деградацією в процесі експлуатації. У відповідності до запропонованої моделі пояснюється виникнення додаткової кількості елементарних дефектів як результат дисоціації трьох видів комплексів точкових дефектів  $(\text{Cu}_i^+ - 2\text{Cu}_{\text{Cd}}^-)^-$ ,  $(\text{V}_{\text{Cd}}^- - \text{Cu}_{i^+})^-$ ,  $(2\text{Cu}_{\text{Cd}}^- - \text{V}_{\text{Te}}^+)^-$ ,  $(\text{Cu}_{i^+} - \text{Cu}_{\text{Cd}}^-)$ . Розглянуто шунтування n-p гетеропереходу і фазові перетворення зі сторони  $p^+ - \text{Cu}_{2-x}\text{Te}$  за рахунок електродифузії  $\text{Cu}_{\text{Cd}}^-$  із p-CdTe на межі n-CdS/p-CdTe та p-CdTe/p<sup>+</sup>-Cu<sub>2-x</sub>Te. З іншого боку, можлива дифузія  $\text{Cu}_i^+$  (межвузлова мідь) в об'єм абсорбера. Можлива електродифузія дефектів із гетеропереходів в об'єм абсорбера, що спричиняє компенсацію ефективних акцепторних центрів та призводить до зниження часу життя неосновних носіїв заряду і відповідно до зниження  $J_f$ . Крім того, спостерігається проростання шунтуючих металевих ланцюжків по поздовжніх межзеренних кордонах p-CdTe між n-p і p-p<sup>+</sup> гетеропереходами та можливість виникнення високоомних фаз системи Cu-Te. Запропонована модель пояснює можливість виникнення фази  $p^+ - \text{Cu}_{2-\delta}\text{S}$  на межі CdS/CdTe, яка стримує проходження фото активної частини сонячного спектру в p-CdTe

**Ключові слова:** телурид кадмію, деградація сонячного елемента, вихідні параметри, світлові діодні характеристики

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# DEGRADATION OF CDTE SC DURING OPERATION: MODELING AND EXPERIMENT

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## 1. Introduction

The increasing use of SC as alternative energy sources is no longer surprising [1]. However, some factors constrain

their widespread use [2]. Among others, particular attention should be paid to the degradation of SC during operation. The rate of degradation of SC based on cadmium telluride can be reduced by optimizing the back contacts to such

instrument structures. Existing approaches to optimizing the back contacts to such instrument structures occur in two directions. The first direction is focused on finding the optimal compound (material) to create the back contact [3]. The second direction is devoted to finding the best way to get the back contact [4].

However, each of these directions requires an understanding of the processes occurring in the solar cell while degradation during operation.

Therefore, it is important to develop a model that will give an understanding of the processes occurring in the solar cell during operation.

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## 2. Literature review and problem statement

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The publications [4–9] present the results of studying the deterioration of characteristics of different types of solar cells. Among those considered, organic [5] solar cells, GaInP/GaInAs/Ge triple-junction solar cells [6, 7], cadmium telluride (CdTe) cells [4, 8], and copper indium gallium selenide cells [9] during operation. Among other types of thin-film solar cells, CdTe solar cells are suitable for both space flight and terrestrial applications due to their economic advantages relative to the production process and high specific power relative to their thickness [10–12]. However, the theoretical value of efficiency for such SC is currently unattainable. One of the reasons for the restrained achievement of the theoretical value of efficiency is the search for optimal contacts to film CdTe SC. For example, in [12] the authors investigated two types of SC: containing graphite paste, as well as Ag and Ni layers as the back contact. As a result of the studies of CdS/CdTe SC, the back contacts made using graphite paste and Ag, as well as graphite paste and Ni, showed similar efficiency values of about 12.5 % and FF at 66 %. In open-circuit conditions, a slight decrease in the efficiency of both SC by 10–15 % was observed.

Thus, the authors concluded that graphite paste behaves as a diffuse barrier preventing the penetration of metal from the secondary contact into the CdTe region. Solar cells using only Ag and Ni layers as the back contacts had significantly lower output FF values (58–62 %) and showed significant degradation. When operating for 707 hours at 100 °C in open-circuit conditions, the efficiency is reduced by 25–45 %. The use of only the Ag layer as the back contact resulted in an increase in the dopant concentration in CdTe due to Ag diffusion into the SC region. Rapid Ag propagation along the grain boundary contributed to shunt formation and increased micro-heterogeneities. Thus, SC having only Ag as the back contact showed a 25 % decrease in efficiency after open-circuit operation. For SC using Ni as the back contact, the formation of the Ni<sub>3</sub>Te<sub>2</sub> intermetallic compound on the CdTe surface was observed. The appearance of such a layer on the CdTe surface contributed to an increase in the series resistance during open-circuit operation of SC for 707 hours at a temperature of 100 °C, resulting in a 45 % efficiency decrease.

The authors of [13] proposed to use the NiTe<sub>2</sub> compound as the back contact to CdS/CdTe SC. The NiTe<sub>2</sub> compound is formed during the chemical deposition of Ni onto the CdTe surface. After holding the obtained SC in the air for 20 hours, no diffusion of Ni into the base layer occurs. The obtained CdS/CdTe SC have an efficiency of more than 10 %, but

their operation in a wet atmosphere revealed a decrease in the output parameters. When introduced into dry air, SC efficiency is restored.

[14] considered the use of the PEDOT-PSS organic back contact, which was applied on the CdTe surface by centrifugation. The efficiency of the resulting solar cell did not exceed 2 %. It was found that such a low efficiency is due to the «through diode» operation of the instrument structure, which is characterized by low open-circuit voltage values.

The authors of [15] investigated the effect of various procedures of etching of CdTe surface, buffer layers and metallization to obtain effective CdS/CdTe SC. Several variants of CdTe surface etching were used: traditional in BrMeOH and NP etching at different concentrations of nitric acid. In addition, solar cells, whose back surface was not chemically etched were investigated. The result is that traditional BrMeOH etching is a relatively mild etching that mainly cleans and creates excess Te on the CdTe surface without significantly expanding the intergranular boundaries. Non-chemically etched SC showed an efficiency of less than 9 %. For such SC, the authors proposed applying a several nm thick layer of elementary Ti to the CdTe surface.

[16] showed that radio-frequency sputtering of Sb<sub>2</sub>Te<sub>3</sub>/Mo onto the CdTe surface is promising for use as a back contact to stable CdTe/CdS SC. This contact is a stable compound having a band gap of about 0.3 eV and low resistivity of about 10<sup>-4</sup> Ohm-cm. Six variants of back contact compositions were investigated, including Sb/Mo, Sb<sub>2</sub>Te<sub>3</sub>/Mo, Cu/Mo, Sb/Au, Sb/Al, Sb<sub>2</sub>Te<sub>3</sub>/Au. After annealing at 150 °C for 30 min, Cu, Al and Au were found to diffuse faster and cause a significant decrease in efficiency. Mo, Sb, and Sb<sub>2</sub>Te<sub>3</sub> diffuse very poorly or do not diffuse at all. The efficiency of SC containing the Sb<sub>2</sub>Te<sub>3</sub>/Mo compound as the back contact increased after annealing by more than 1 %. This indicates that the conditions of back contact formation were not optimal. Further annealing to 400 °C in the air did not change the efficiency.

Long-term stability tests were performed at a constant illumination of about 1,000 W/m<sup>2</sup> at 80 °C, which corresponds to a 100-fold acceleration of lifetime [17]. SC, whose back contacts were made using Sb and Sb<sub>2</sub>Te<sub>3</sub> as the buffer layer and Mo as the secondary contact were resistant to degradation. Whereas SC with Au or Al metallization degraded faster. The height of the potential barrier of SC having the Sb<sub>2</sub>Te<sub>3</sub>/Mo back contact is lower than that of SC containing Sb/Mo as the back contact. However, the use of Sb/Mo is economical for industrial production.

In [18, 19], SC with the copper-containing back contact were investigated. It is shown that during the SC operation, the fill factor of the light current-voltage characteristic is reduced due to shunting effects. Therefore, on the one hand, to obtain stable CdS/CdTe solar cells, it is necessary to avoid using Cu in the back contact. On the other hand, Cu-containing CdS/CdTe SC remain versatile because, regardless of the base layer formation technology, they provide high efficiency of photovoltaic processes. At the same time, the most optimal etchant creating the elementary Ti layer is BrMeOH. Back contacts that do not contain Cu either do not provide highly efficient instrument structures, or are combined with a specific base layer technology. Thus, in order to create effective SC for large-scale production, it is necessary to understand the processes occurring in SC during operation that lead to the degradation of the output parameters.

### 3. The aim and objectives of the study

The aim of the work is to develop a model of CdS/CdTe SC degradation during operation to further develop a method to restore the effectiveness of such SC.

To achieve this aim, the following objectives are set:

- to investigate the output parameters and diode characteristics of SC after accelerated degradation;
- to investigate the degradation mechanisms of film cadmium sulfide and telluride SC with copper back contacts.

### 4. Materials and methods for CdTe SC research

The production of 10×10 cm experimental specimens of SC was carried out according to the standard procedure for CdTe, i. e., by thermal vacuum evaporation using a UVN67 vacuum unit with modified internal equipment. [20] The thickness of the CdTe base layer was 2.5 μm. The internal equipment of the unit is shown in Fig. 1.



Fig. 1. Inner structure of the CdS and CdTe sputtering unit: 1, 2 – screens; 3 – cadmium telluride powder evaporator; 4 – cadmium sulfide powder evaporator; 5 – carousel, 6 – substrate heater, 7 – substrate holder [20]

The next operation was chloride treatment and annealing in air at 200 °C for 30 minutes. During the previous 10 minutes, the laboratory specimens were heated to the specified annealing temperature.

The application of ITO films (indium and tin oxides) was carried out by non-reactive DC magnetron sputtering in a VUP-5M vacuum unit (Fig. 2).

Since effective instrument structures cannot be obtained without the copper layer, a 2 nm nanoscale copper layer was deposited on the cadmium telluride surface prior to ITO application. Thickness minimization of the copper layer was aimed at increasing the degradation resistance of the instrument structure. The structure of the studied specimens is presented in Fig. 3.

After manufacture, SC were investigated in standard laboratory conditions, adopted for testing SC for ground use, i. e. in the AM1.5 mode at a luminous power of 100 mW/cm<sup>2</sup> and temperature of 25 °C. Their light CVC were measured, followed by dark voltage-capacitance characteristics (VCC) at 20–22 °C. To study the degradation processes in such SC, after measuring the initial parameters, SC were placed in a sealed plastic box and held for 4 years at 15–25 °C. After holding under these conditions, the SC were removed and measurements of light CVC and dark VCC were repeated.

Transmission and reflection spectra of cadmium telluride films obtained on glass substrates were also investigated. This allowed determining the spectral dependence of the absorption coefficient of cadmium telluride films. The absorption intensity of the solar spectrum in the base layer was calculated by integrating the absorption coefficient value in the spectral interval corresponding to solar radiation in ground conditions.



a



b

Fig. 2. VUP-5M vacuum unit: a – vacuum unit; b – material-saving magnetron [21]

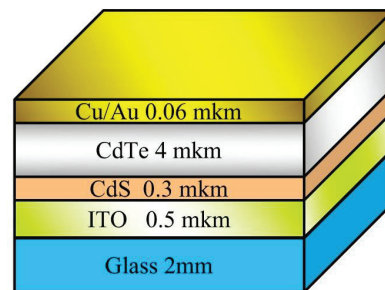


Fig. 3. Structure of ITO/CdS/CdTe/Cu/Au solar cells

### 5. Results of the study of CdS/CdTe SC after long-term storage

#### 5.1. Results of the study of the output parameters and diode characteristics of SC before and after accelerated degradation

As a result of the CdTe SC study, the output parameters and diode characteristics of the instrument structures were determined both immediately after manufacture and after



holding for 4 years at a temperature of 15–25 °C. Light CVC of the studied specimens are shown in Fig. 4.

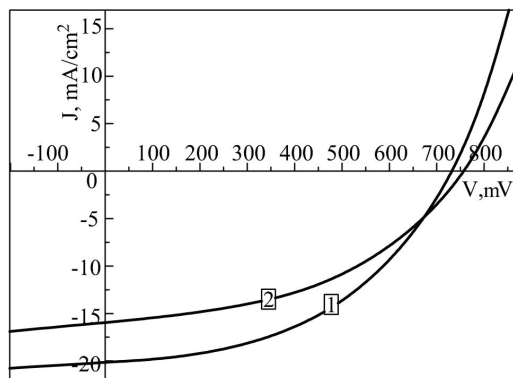


Fig. 4. Light CVC of ITO/CdS/CdTe/Cu/Au SC: 1 – after manufacture; 2 – after long-term storage

The obtained output parameters and diode characteristics of CdS/CdTe SC calculated using the method given in [17] before and after long-term storage are given in Table 1.

Table 1

Changes in the output parameters and diode characteristics of CdS/CdTe SC after long-term storage

Parameters and characteristics	After manufacture	After long-term storage
$J_{PH}$ , mA/cm <sup>2</sup>	21.3	16.9
$J_0$ , 10 <sup>-7</sup> A/cm <sup>2</sup>	1.9·10 <sup>-7</sup>	1.5·10 <sup>-5</sup>
$R_s$ , Ohm·cm <sup>2</sup>	1.6	15.4
$R_{sh}$ , Ohm·cm <sup>2</sup>	623	750
$J_{sc}$ , mA/cm <sup>2</sup>	21.2	16.0
$V_{oc}$ , mV	731	756
$FF$ , c. u.	0.66	0.62
Efficiency, %	10.2	5.8

### 5. 2. Results of the study of spectral dependences of SC after accelerated degradation

After long-term storage, spectral dependences of the studied instrument structures were obtained. A typical spectral dependence for the ITO/CdS/CdTe/Cu/Au specimens is shown in Fig. 5.

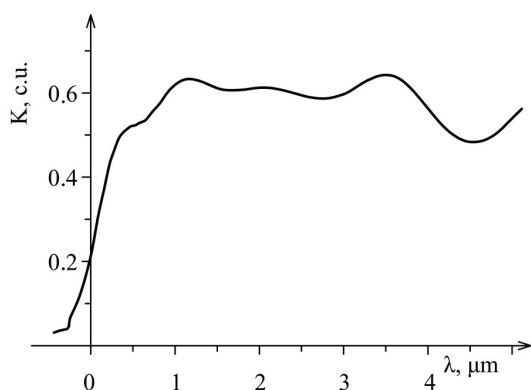


Fig. 5. Typical transmission spectrum of cadmium telluride films. Investigated instrument structures ITO/CdS/CdTe/Cu/Au

The study of the spectral dependences of the transmittance showed that the ITO/CdS/CdTe/Cu/Au instrument structures in the spectral range (0.82–1.20) μm have an average transmittance of 0.58.

## 6. Discussion of the results of the study of CdS/CdTe SC after long-term storage

### 6. 1. Results of the study of degradation mechanisms of film SC based on cadmium sulfide and telluride with copper-containing back contacts

Analysis of Table 1 shows that the main contribution to the degradation process of CdS/CdTe SC with the copper-containing back contact is made by such diode characteristics as  $J_{ph}$  and  $R_s$ , while changes in these parameters lead to a significant decrease in  $R_{sh}$ . In turn, given the relations described in [18], this leads to a significant drop in the output parameters  $J_{sc}$ ,  $V_{oc}$ ,  $FF$ , and hence the efficiency. Earlier in [21, 22], the influence of such complexes of point defects as  $Cu_{Cd}^-$ ,  $Cu_i^+$ ,  $V_{Cd_s}^+$ ,  $(Cu_i^+ - Cu_{Cd})$ ,  $(Cu_i^+ - 2Cu_{Cd}^-)$ ,  $(V_{Cd_s}^+ - Cu_i^+)$ ,  $(2Cu_{Cd_s}^- - V_{Te}^+)$  and their elementary components on the parameters of SC containing copper in the back contact was determined. In view of this, the key mechanisms of degradation of the investigated SC during long-term storage, acting solely at the expense of the internal resources of the SC heterostructure, are probably as follows. First of all, the decisive role of thermodynamic mass transfer of copper from the region containing copper telluride (arising from the back contact at the stage of SC production) through the absorber along the boundaries and through the volume of CdTe grains to CdS, as well as the diffusion interaction of CdS and CdTe layers is undoubted. The validity of this assumption is confirmed experimentally in [21, 22], where an understanding of a number of important mechanisms of the effect of structural transformations in the semiconductor base of SC on the evolution of electronic, added and output parameters was also achieved.

Thus, it can be concluded that the decisive role in the SC degradation during operation is played by copper diffusion from the back contact through the absorber along the grain boundaries and through their volume at the stage of SC production. The change in the height of the potential barrier to  $F_{np} = 2.4$  eV revealed in this study may indicate the dissolution of CdS and CdTe.

However, in the previously published papers [14, 18–20], the role of two space charge regions with the built-in electric field remains out of view. Considering the effect of the built-in electric field of the  $n-p$  heterojunction on the SC parameters during operation, it should be taken into account that in this region, after the SC production, there is more copper than in the absorber region [23, 24]. In addition, there is a diffusion of  $CuCd^-$  from the absorber to the  $n$ -CdS layer and  $Cu_i^+$ , on the contrary, to the absorber volume. As a result of  $CuCd^-$  diffusion from the absorber region to the  $n$ -CdS layer, at the CdS/CdTe boundary, the  $p^+ - Cu_{2-\delta}S$  phase and  $CuCd^-$  centers may occur, which may lead to an increase in  $R_s$ . There may also be an expansion of the depletion region from  $n$ -CdS, which leads to both an increase in  $R_s$  and a decrease in  $J_{ph}$ ,  $J_{sc}$  and  $V_{oc}$ . The bandgap  $p^+ - Cu_{2-\delta}S$  depending on  $\delta$  is 1.0–2.3 eV, which is less than  $E_g = 2.42$  eV in CdS. In view of this, as well as the fact that  $Cu_{2-\delta}S$  is a direct-gap semiconductor, the partial decrease of  $J_{ph}$ , and therefore  $J_{sc}$ ,  $V_{oc}$  and  $FF$ , during the SC storage can be explained by the

decrease in the flux of photovoltaic active light quanta entering the absorber due to their additional absorption in  $p^+-Cu_{2-\delta}S$ . Note that an additional factor contributing to the formation of  $p^+-Cu_{2-\delta}S$  may be the effect of partial self-compensation in  $n$ -CdS:Cu<sub>Cd</sub><sup>-</sup>, which leads to the generation of sulfur vacancies, resulting in an excessive amount of sulfur at the  $n$ -CdS and CdTe boundary capable of actively reacting with excess copper present there.

### 6. 2. Development of a model of CdS/CdTe SC degradation during operation

Comparison of the output parameters and light-emitting diode characteristics with those after degradation, supplemented by the results of voltage-capacitance characteristics, allowed proposing the following degradation model. The model takes into account all the considered processes of mass transfer, phase transformations and their consequences on the evolution of photocurrent, electronic, diode characteristics and output parameters of SC. The driving force for the diffusion of Cu into CdS and S into Cu<sub>2-x</sub>Te along the grain boundaries is the large Cu-S bond strength compared to Cu-Te.

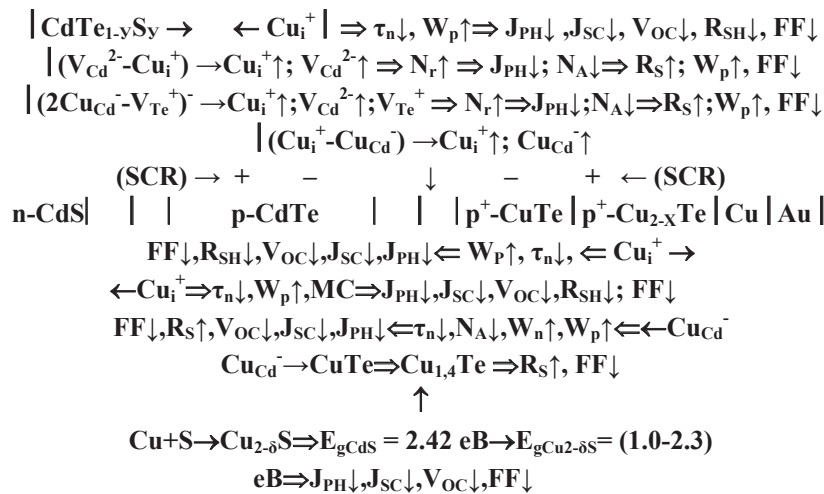


Fig. 6. Model of SC degradation:

- $\tau_n$  – electron lifetime in  $p$ -CdTe;  $W_p$  – width of the depletion region of  $p$ -CdTe;
- $N_r$  – volume concentration of recombination centers in  $p$ -CdTe;
- $N_A$  – concentration of active acceptor centers in  $p$ -CdTe;  $W_n$  – width of the depletion region of the  $n$ -CdS layer; SCR – space charge region

The resulting physical understanding of the mechanisms of degradation of film SC based on sulfide and cadmium telluride during operation will allow developing a method to restore the efficiency of ITO/CdS/CdTe/Cu/Au SC after their degradation. These results can be used for sulfide and cadmium telluride SC with the Cu-containing back contact.

### 7. Conclusions

1. The study of the output parameters and diode characteristics of CdS/CdTe SC with the copper back contact after accelerated degradation is performed. It is found that the main contribution to the process of degradation of CdS/CdTe SC with the copper-containing back contact is by such output parameters and diode characteristics as  $J_{ph}$  and  $R_s$ , while a significant decrease in  $R_{sh}$  is observed.

2. The model of degradation and change of the parameters of  $n^+$ -ITO/ $n$ -CdS/ $p$ -CdTe/ $p^+-Cu_{2-x}Te$ /Cu/Au SC is developed. According to the proposed model, the occurrence of additional elementary defects as a result of dissociation of three types of point defect complexes  $(Cu_i^+ - 2Cu_{Cd}^-)$ ,  $(V_{Cd}^{2-} - Cu_i^+)$ ,  $(2Cu_{Cd}^- - V_{Te}^+)$  is explained. Shunting of the  $n$ - $p$  heterojunction and phase transformations from the  $p^+-Cu_{2-x}Te$  side due to electrodiffusion of  $Cu_{Cd}^-$  with  $p$ -CdTe at the boundary of  $n$ -CdS/ $p$ -CdTe and  $-CdTe/p^+-Cu_{2-x}Te$  is considered. On the other hand,  $Cu_i^+$  (interstitial copper) diffusion into the absorber volume is possible. Thus, electrodiffusion of defects from heterojunctions into the absorber volume is possible, which results in compensation of effective acceptor centers and leads to a decrease in the lifetime of minority charge carriers and consequently a decrease in  $J_{ph}$ . In addition, there is a growth of shunting metal chains along the longitudinal intergranular boundaries of  $p$ -CdTe between  $n$ - $r$  and  $p$ - $p^+$  heterojunctions and the possibility of high-resistance phases of the Cu-Te system. The proposed model explains the possibility of the occurrence of the  $p^+-Cu_{2-\delta}S$  phase at the CdS/CdTe boundary, which constrains the passage of the photoactive part of the solar spectrum in  $p$ -CdTe.

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