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# Preparation and Investigation of p-GaAs/n-Cd<sub>1-x</sub>Zn<sub>x</sub>S<sub>1-y</sub>Te<sub>y</sub> Heterojunctions Deposited by Electrochemical Deposition

**Huseyn M. Mamedov**

Faculty of Physics,  
Department of Physical Electronics,  
Baku State University,  
Z.Khalilov street, 23,  
Baku Az1148, Azerbaijan  
e-mail: mhhuseyng@gmail.com

**Zoltan Konya**

Department of Applied and  
Environmental Chemistry,  
University of Szeged,  
H-6720 Szeged, Rerrich Bela ter 1.,  
Hungary  
e-mail: konya@chem.u-szeged.hu

**Mustafa B. Muradov**

Faculty of Physics,  
Nanomaterials Laboratory,  
Baku State University,  
Z.Khalilov street, 23,  
Baku Az1148, Azerbaijan  
e-mail: mbmuradov@gmail.com

**Akos Kukovecz**

Department of Applied and  
Environmental Chemistry,  
University of Szeged,  
H-6720 Szeged, Rerrich Bela ter 1.,  
Hungary  
e-mail: kakos@chem.u-szeged.hu

**Krisztian Kordas**

Microelectronics and Materials  
Physics Laboratories,  
University of Oulu,  
P.O. Box 8000FI-90014, Oulu, Finland  
e-mail: lapy@ee.oulu.fi

**Daniel P. Hashim**

Department of Mechanical Engineering and  
Materials Science,  
Rice University,  
6100 Main Street, MS-321,  
Houston, TX 77005  
e-mail: danielpaul3@gmail.com

**Vusal U. Mamedov**

Faculty of Physics,  
Department of Physical Electronics,

Baku State University,  
Z.Khalilov street, 23,  
Baku Az1148, Azerbaijan  
e-mail: mammadovv@gmail.com

*Anisotype heterojunctions of p-GaAs/n-Cd<sub>1-x</sub>Zn<sub>x</sub>S<sub>1-y</sub>Te<sub>y</sub> have been fabricated by preparing n-type Cd<sub>1-x</sub>Zn<sub>x</sub>S<sub>1-y</sub>Te<sub>y</sub> thin films onto p-GaAs single crystal wafers using an electrochemical deposition method. The voltammetric behavior of the Cd<sub>1-x</sub>Zn<sub>x</sub>S<sub>1-y</sub>Te<sub>y</sub> thin films on GaAs substrates from aqueous solutions was studied. Electrical and photoelectrical properties of heterojunctions were studied depending on the Cd<sub>1-x</sub>Zn<sub>x</sub>S<sub>1-y</sub>Te<sub>y</sub> films composition ( $x = 0.1 \div 0.8$ ;  $y = 0.2; 0.4; 0.9$ ) and heat treatment (HT) regime in argon atmosphere (100–450 °C during 3–16 min). Under AML5 conditions, the open-circuit voltage, short-circuit current, fill factor, and efficiency of our best cell, was  $V_{oc} = 584$  mV,  $J_{sc} = 14.54$  mA/cm<sup>2</sup>,  $FF = 0.6$ , and  $\eta = 6.7\%$ , respectively.*  
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*Keywords: electrochemical deposition, thin film, heterojunction, heat treatment, solar cell*

## Introduction

Thin films of II-VI compounds (CdS, CdTe, Cd<sub>1-x</sub>Zn<sub>x</sub>S, Cd<sub>1-x</sub>Zn<sub>x</sub>S<sub>1-y</sub>Se<sub>y</sub>, and Cd<sub>1-x</sub>Zn<sub>x</sub>S<sub>1-y</sub>Te<sub>y</sub> (CZSTE), etc.) have attracted considerable attention from the research community due to their wide uses in the fabrication of semiconductor device technology and solar cells [1–5]. In photovoltaic systems, the replacement of CdS with the higher energy band gap of Cd<sub>1-x</sub>Zn<sub>x</sub>S, Cd<sub>1-x</sub>Zn<sub>x</sub>S<sub>1-y</sub>Se<sub>y</sub>, and CZSTE alloys has led to a decrease in window absorption losses and has resulted in an increase in the short-circuit current. The II-VI quaternary semiconductors seem to be useful materials with photosensitivity in the visible and ultraviolet wavelength regions [6–10]. Since single crystals of GaAs are well-studied materials, their use at manufacturing of heterojunctions p-GaAs/CZSTE will be a good way to deeply study the physical properties of CZSTE films.

There are many techniques used to synthesize thin films of II-VI compounds, such as thermal evaporation, chemical bath deposition, successive ionic layer absorption and reaction, magnetron sputtering, metalorganic vapor phase epitaxy, etc. [11–17]. In photovoltaic applications, where semiconductor films over large areas are required, the electrodeposition technique is specially adequate. In addition, for application in solar cells, electrodeposition allows one to easily alter both the bandgap and lattice constant by composition modulation through the control of growth parameters such as applied potential, pH, and temperature of the bath [11,18–20]. Thus, it is at least in principle possible to easily grow large areas of tandem cells designed for the most efficient conversion of the solar spectrum.

In the present work, anisotype heterojunctions of p-GaAs/n-CZSTE were fabricated by depositing CZSTE thin films as a window using the electrochemical deposition method onto the p-GaAs single crystals.

## Experimental

Electrodeposition of the CZSTE films onto the p-GaAs substrates was carried out at a temperature of 80 °C from aqueous solution containing cadmium (CdSO<sub>4</sub>), zinc (ZnSO<sub>4</sub>), sodium (Na<sub>2</sub>S<sub>2</sub>O<sub>3</sub>), and tellurium (TeO<sub>2</sub> or Na<sub>2</sub>Te<sub>2</sub>O<sub>3</sub>) salts. The thickness and resistivity of the monocrystalline p-GaAs substrates were 0.4 mm and  $\rho = 0.2\text{--}0.23$  Ω cm, respectively. Before the deposition process, the surfaces of the GaAs substrates were etched in an aqueous solution of hydrochloric acid and KOH-KNO<sub>3</sub> (1:3) composition for 3 min. After etching, the GaAs wafers were washed for 2 min in pure alcohol and distilled water maintained at high temperatures ( $\geq 300$  °C).

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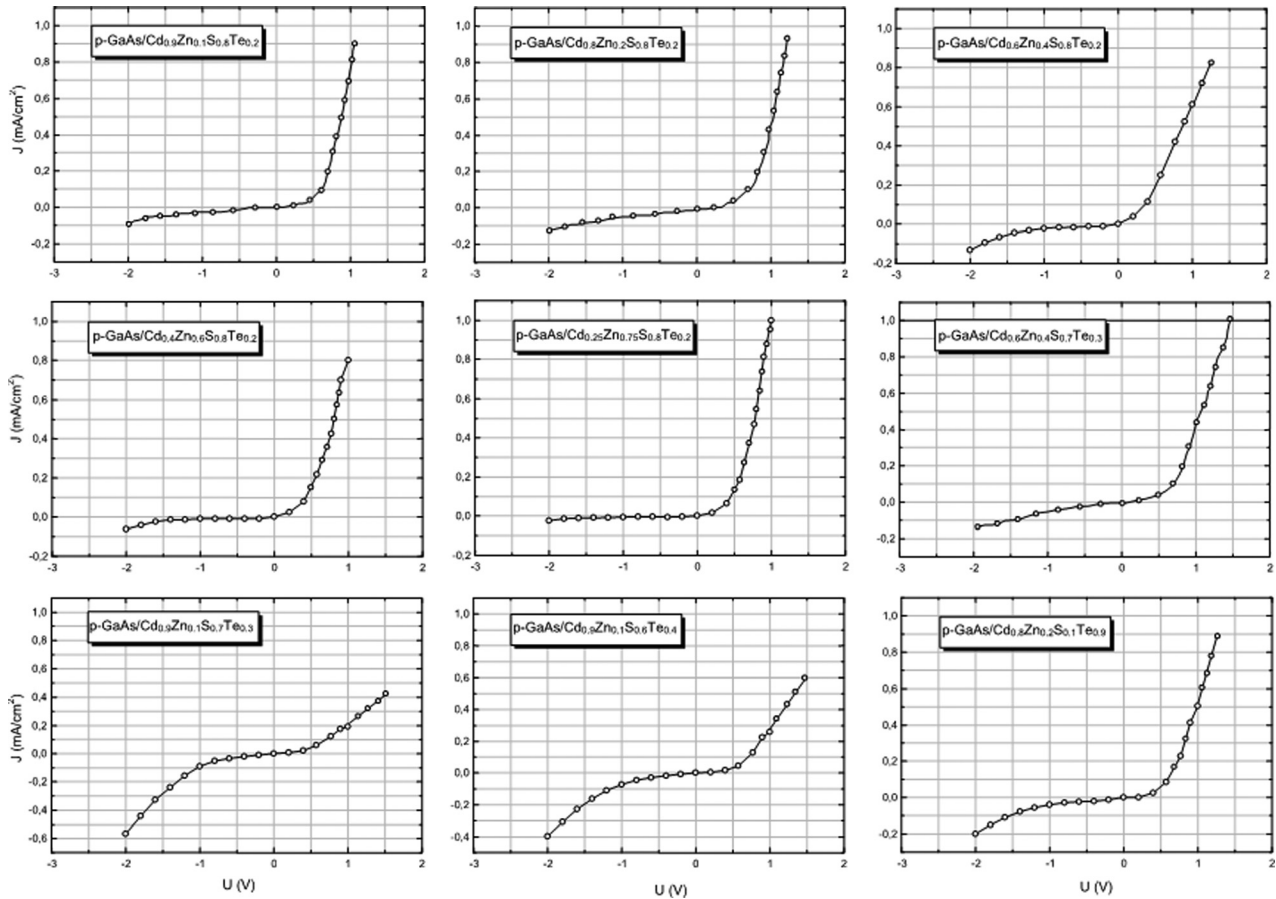


Fig. 1 Dark  $J$ - $V$  curves for as-deposited p-GaAs/CZSTE heterojunction

112 Cyclic voltammetry was used to monitor the electrochemical  
 113 reactions in separate solutions of  $\text{CdSO}_4$ ,  $\text{ZnSO}_4$ ,  $\text{Na}_2\text{S}_2\text{O}_3$ , and  
 114  $\text{TeO}_2$ , then in their combined solution at the same concentration  
 115 and  $p\text{H}$ . The cyclic voltammogram was scanned in the potential  
 116 range  $1.2\text{ V}$  to  $-1.2\text{ V}$  versus graphite (or  $\text{Ag}/\text{AgCl}$ ) electrodes.  
 117 Cyclic voltammogram for mixture of  $\text{CdCl}_2$ ,  $\text{ZnCl}_2$ ,  $\text{Na}_2\text{S}_2\text{O}_3$ , and  
 118  $\text{Na}_2\text{Se}_2\text{O}_3$  salts shows that wave  $-0.52 \div -0.9\text{ V}$  corresponded to  
 119 the formation of CZSTE layers. The thickness of the CZSTE films  
 120 grown by electrodeposition from a solution could be varied in a  
 121 wide range from  $50$  to  $1600\text{ nm}$ .

122 In order to fabricate the heterojunctions, an ohmic in electrode,  
 123 in reticulate form was evaporated on the CZSTE films with an  
 124 area of  $\sim 0.82\text{--}1\text{ cm}^2$ . An ohmic contact was performed on the  
 125 side of GaAs wafers by evaporating an Al electrode.

## 126 Results and Discussion

127 The dark current-voltage ( $J$ - $V$ ) curves of the heterojunctions  
 128 were measured in the direct and reverse current modes. The experi-  
 129 mental  $J$ - $V$  curves, measured at  $300\text{ K}$ , for as-deposited p-GaAs/  
 130 CZSTE heterojunctions, using various values of  $x$  and  $y$ , are illus-  
 131 trated in Fig. 1.

132 These curves definitely proved diode type behavior, with the  
 133 forward direction corresponding to the positive potential on  
 134 p-GaAs. Thus, according to this figure, the as-deposited junctions  
 135 composed of CZSTE films with  $x=0.75$  and  $y=0.2$  (which is a  
 136 good lattice match with GaAs layers) reaches a rectification value  
 137 of  $k=700$  at voltage  $U=1.0\text{ V}$  ( $k$  is the rectification factor), and  
 138 decreases when zinc concentration,  $x$ , increases. The low rectifica-  
 139 tion coefficient is due to the high series resistance within the hetero-  
 140 structure. Plotting the natural log of the current density versus  
 141 the applied voltage, we are able to identify a characteristic thermally  
 142 activated recombination region up to  $0.63\text{ V}$ . Usually, such  
 143 dependencies are described by the expression

$$J = J_s \left[ \exp\left(\frac{eV}{AkT}\right) - 1 \right] \quad (1)$$

Here,  $J_s$  is the saturation current density,  $V$  is the applied voltage,  $e$  is the electron charge,  $A$  is the ideality factor,  $k$  is the Boltzmann constant, and  $T$  is the temperature.

Increasing the forward bias magnitude ( $U > 0.65\text{ V}$ ) resulted in a less steep dependence of  $J(V)$  and its pronounced deviation from the curve calculated according to the formula (1), which can be a consequence of the changes of carrier transport mechanism. The most possible case to be considered is tunneling recombination. In the as-deposited heterojunctions, the ideality factor was determined under a forward bias, and it was normally found to range from  $1.6$  to  $2.7$  for the different  $x$  and  $y$ . This established that the value of ideality factor was minimal for the p-GaAs/n- $\text{Cd}_{0.25}\text{Zn}_{0.75}\text{S}_{0.8}\text{Te}_{0.2}$  heterojunctions.

The mechanism of current passage through the heterojunctions essentially changes with increasing HT temperature from  $0$  to  $390\text{ }^\circ\text{C}$  (for  $14\text{ min}$ ). Notably, tunnel currents sharply decreased with increasing HT temperature, which testifies to reduction of defects and decreasing series resistance (Table 1). After the HT in argon atmosphere at  $390\text{ }^\circ\text{C}$  for  $14\text{ min}$ , the ideality factor values were approximately  $1.4$  for the heterojunctions with  $x=0.75$  and  $y=0.2$ . It is significant to note that the best rectification for the annealed p-GaAs/n- $\text{Cd}_{0.25}\text{Zn}_{0.75}\text{S}_{0.8}\text{Te}_{0.2}$  heterojunctions was obtained at about  $k=3000$ , which is attributable to the optimal HT conditions and lattice mismatch between the solid solution of  $\text{Cd}_{0.25}\text{Zn}_{0.75}\text{S}_{0.8}\text{Te}_{0.2}$  and GaAs.

The capacitance versus voltage measurement results ( $1/C^2$ - $V$ ) for the heterojunctions p-GaAs/n- $\text{Cd}_{0.25}\text{Zn}_{0.75}\text{S}_{0.8}\text{Te}_{0.2}$  annealed in argon atmosphere at  $390\text{ }^\circ\text{C}$  for  $14\text{ min}$  showed a linear relationship with bias voltage and indicates that the junction is abrupt.

AQ3

AQ4

Table 1

HT temperature and duration	Rectification coefficient ( $k$ )	Nonideality factor ( $A$ )	Series resistance ( $R_s, \Omega \text{ cm}^2$ )
before HT	200	1.61	260
150 °C; 14 min	540	1.54	200
200 °C; 14 min	970	1.51	176
250 °C; 14 min	1700	1.46	93
300 °C; 14 min	2450	1.44	54
350 °C; 14 min	2600	1.42	30
390 °C; 14 min	3000	1.4	24
430 °C; 14 min	6	2.56	1300

173 Also built-in potential ( $V_{bi} = 0.61 \text{ V}$ ) were calculated by extrapolating ( $1/C^2 - V$ ) plot to ( $1/C^2 = 0$ ).

175 As-deposited (nonheat-treated)  $p$ -GaAs/CZSTE heterojunctions were found to possess a photovoltaic effect. As follows from Fig. 2, the efficiency of the heterojunctions depends on the film's composition  $x$  and  $y$ . Under AM1.5 conditions, the maximal values of open-circuit voltage, short-circuit current, fill factor, and efficiency for cells  $p$ -GaAs/ $n$ - $\text{Cd}_{0.25}\text{Zn}_{0.75}\text{S}_{0.8}\text{Te}_{0.2}$ , were  $V_{oc} = 131 \text{ mV}$ ,  $J_{sc} = 3.4 \text{ mA/cm}^2$ ,  $\text{FF} = 0.43$ , and  $\eta = 0.2\%$ , respectively.

183 To assess the effect of HT on the photoelectric properties of the heterojunctions, the films were annealed in argon atmosphere at 100–450 °C for 3–16 min. Figure 3 shows typical spectral dependences of the photocurrent for  $p$ -GaAs/ $n$ - $\text{Cd}_{0.25}\text{Zn}_{0.75}\text{S}_{0.8}\text{Te}_{0.2}$  heterojunctions before and after HT. There occurs a reconstruction of the photosensitivity spectrum after HT, i.e., the spectrum broadens. As the HT temperature increased from 0 to 390 °C for 14 min, photosensitivity in the  $\lambda_m = 0.38\text{--}0.8 \mu\text{m}$  wavelength region sharply increased. The near infrared photosensitivity falloff for all heterojunctions indicated GaAs absorber band gaps of 1.42 eV. Figure 3 also shows that after subsequently HT in argon atmosphere for 14 min at  $\geq 400 \text{ °C}$  the performance of these cells deteriorated.

196 The observed effect of HT on the heterojunction properties can be understood in terms of electronic–molecular interaction between the surface of CZSTE films and oxygen [3–5]. It is believed that oxygen adsorption, after the removal of CZSTE films from the solution, leads to the formation of deep acceptor states in the surface layer of the films. The oxygen-related acceptors

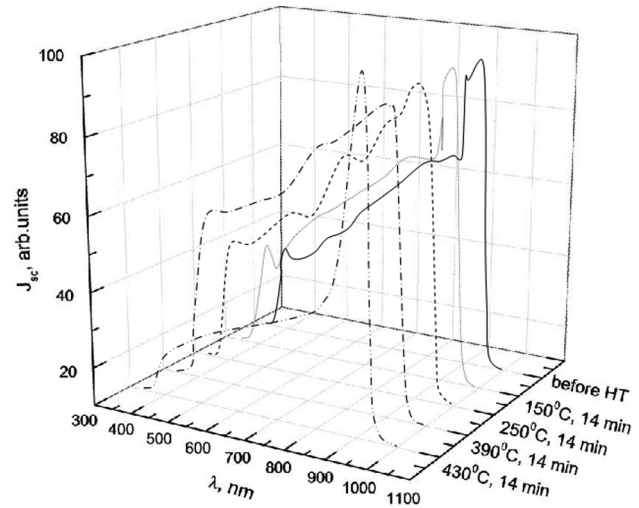


Fig. 3 Spectral dependences of the photocurrent for  $p$ -GaAs/ $n$ - $\text{Cd}_{0.25}\text{Zn}_{0.75}\text{S}_{0.8}\text{Te}_{0.2}$  heterojunctions before and after HT

capture electrons from the film bulk, creating a near-surface potential barrier, which is responsible for the low short-wavelength photosensitivity of the nonheat-treated heterojunctions. The small height of the intergranular barriers in polycrystalline films, as compared to the oxygen-related barriers, renders the short-wavelength photoresponse of the  $p$ -GaAs/CZSTE heterojunctions to be governed by the density of oxygen-related states. The observed effect of HT on the photoelectric properties of the heterojunctions demonstrates that the donor and acceptor concentrations in the films depend on HT conditions. In particular, it seems likely that, in the initial stages of HT, some of the oxygen desorbs, which enhances the short-wavelength photosensitivity of the heterojunctions. In addition, HT at 390 °C for 14 min results in preferential vaporization of Cd and Zn. The Cd and Zn vacancies forming in the surface layer of the CZSTE films act as  $r$  centers. The decrease in the density of surface defects and film recrystallization during subsequent HT shifts the photosensitivity maximum to shorter wavelengths and improves the performance parameters of the films. The sharp decrease in photosensitivity upon heat treatment at 400 °C or higher temperatures indicates that some of

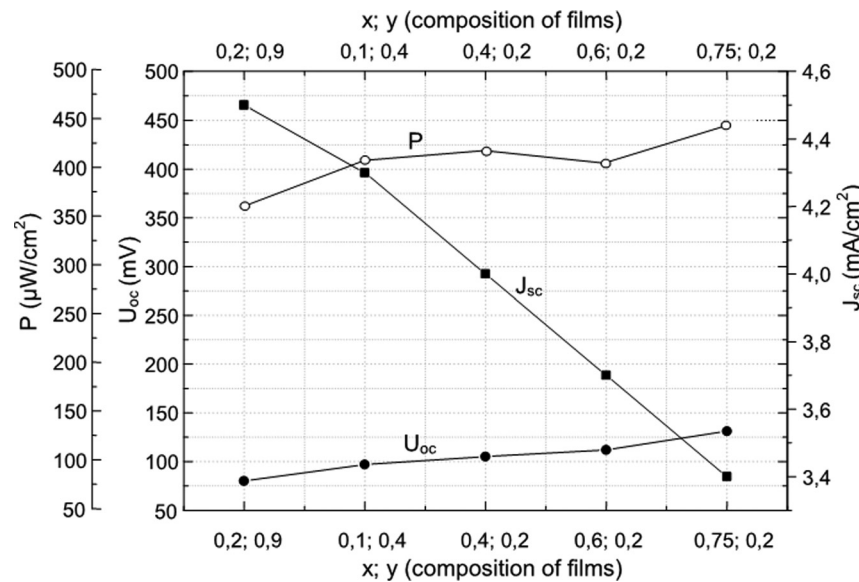
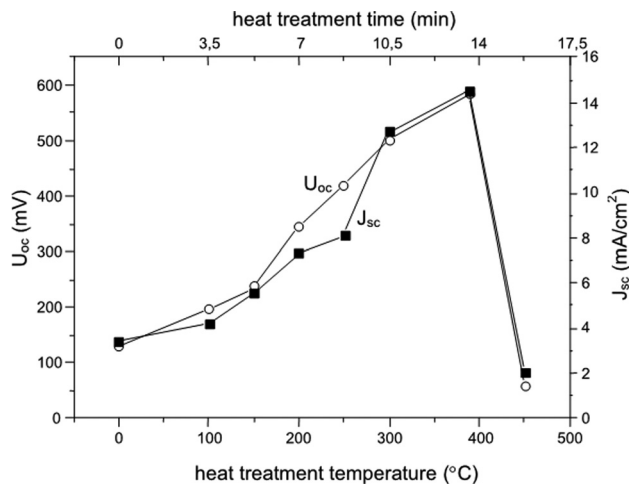


Fig. 2 Dependence of short-circuit current ( $J_{sc}$ ), open-circuit voltage ( $U_{oc}$ ), and power output ( $P$ ) of the as-deposited  $p$ -GaAs/ $n$ - $\text{Cd}_{1-x}\text{Zn}_x\text{S}_{1-y}\text{Te}_y$  cells on the films composition



**Fig. 4** Dependence of p-GaAs/n-Cd<sub>0.25</sub>Zn<sub>0.75</sub>S<sub>0.8</sub>Te<sub>0.2</sub> solar cell parameters on the heat treatment time and temperature

the oxygen does not desorb and remains in the surface layer in atomic form. As a result, the  $r$  centers begin to play a crucial role in determining the recombination process, and the concentration of holes captured by the  $r$  centers increases sharply, reducing the photoresponse of the devices.

Note that, under the conditions of this study, the short-circuit current through the heterojunctions varies nonmonotonically not only with temperature but also with HT time and reaches a maximum after heat treatment at 390 °C for 14 min (Fig. 4). Under AM1.5 conditions the maximal values of open-circuit voltage, short-circuit current, fill factor and efficiency of our best cell, were  $V_{oc} = 584$  mV,  $J_{sc} = 14.54$  mA/cm<sup>2</sup>, FF = 0.6, and  $\eta = 6.7\%$ , respectively.

During storage for more than 36 months at room temperature, the parameters of HT p-GaAs/CZSTE heterojunctions experienced no degradation.

## 238 Conclusions

p-GaAs/CZSTE heterojunctions prepared by the method of electrochemical deposition are suitable to fabricate high efficiency solar cells. Their electrical and photoelectrical characteristics were studied depending on the composition of CZSTE films and the HT condition. It is established that HT at 390 °C for 14 min in argon atmosphere reduces the concentration of defects, results in formation of heterojunctions and minimum values of nonideality factor ( $A = 1.4$ ) of  $J$ - $V$  characteristics and serious resistance ( $R_a = 24 \Omega \text{ cm}^2$ ). The forward current of this junction obeys tunneling-recombination model and ( $C$ - $V$ ) measurements revealed that heterojunctions are abrupt.

Heterojunctions with  $x = 0.75$  and  $y = 0.2$  possess a high photosensitivity after the HT in argon at 390 °C for 14 min. Under standard 100 mW/cm<sup>2</sup> white-light illumination at room temperature, the values of the parameters of our best cell were

$V_{oc} = 584$  mV,  $J_{sc} = 14.54$  mA/cm<sup>2</sup>, FF = 0.6, and  $\eta = 6.7\%$ , respectively.

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