# Organic-inorganic hybrid tandem bulk heterojunction ITO/AIPc:H<sub>2</sub>Pc/n-Si/Al photoelectric cell

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Organic-inorganic hybrid tandem heterojunction ITO/AIPc:H<sub>2</sub>Pc/n-Si/AI photoelectric cell was fabricated by a pressing technology using preliminary vapor deposited bulk heterojunction films of mixed aluminum-phthalocyanine (AIPc) and metal free phthalocyanine (H<sub>2</sub>Pc) on the n-Si substrate and ITO coated plastic substrate. By keeping face to face the heterojunction films, the substrates (silicon and ITO coated plastic substrates) were pressed and fixed by adhesive at elevated temperature conditions. Total thickness of the AIPc and H<sub>2</sub>Pc films were equal to 340 nm. On the back side of n-Si substrate the AI film was deposited. Device architecture was the following: ITO/AIPc:H<sub>2</sub>Pc/n-Si/AI. The morphology of the organic semiconductors film was investigated by AFM. The optical properties of the AIPc-H<sub>2</sub>Pc film were studied by UV-visible spectroscopy. Current–Voltage characteristics were measured at dark condition and under illumination as well. Under illumination of 290 W/m<sup>2</sup> the values of V<sub>oc</sub>, I<sub>SC</sub>, *FF* and efficiency were equal to 0.32 V, 0.9 mA/cm<sup>2</sup>, 0.37 and 0.22 %, respectively.

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### 1. Introduction

Among all kinds of energy electric power plays a crucial role in domestic and industrial applications. It is predicted that the contribution of photovoltaic systems in the world energy consumption will be equal to around 4 % Ideally, cost of electric energy should be in 2020. acceptable for users, materials and technology of device fabrication should be environmentally friendly, and recycling of the devices should be efficient [1-4]. These problems can be resolved if organic solar cell technologies will be developed and implemented in practice [5]. One of the important point concerning the technology of organic solar cells (OSC) is the relatively cheap film deposition methods used [6-8]: spin-coating deposition of organic semiconductors from solution, doctor blading, screen printing and vacuum deposition at lower temperatures, 400-600°C, are some of the examples. At present ink-jet printing technology is developing as well for deposition of thin films [8-11]. Moreover, the organic materials because of their large surface area, flexibility, low density, easy availability, low cost and easy processing [12-14] have become fascinating for researchers to investigate their talent to replace the traditional inorganic materials in electronic devices [15]. From last few decades remarkable research work is being carried out on a variety of organic

materials [16] and some of organic materials based electronic devices such as sensors and light emitting diodes (OLEDs) have been commercialized [17, 18]. Out of organic materials phthalocyanines and their derivatives are non-toxic, thermally and chemically stable and easy to process p-type semiconductors. These properties make the phthalocyanine very suitable for electronic and optical devices [15, 19]. The phthalocyanine are being investigated for their applications in OFETs, OLEDs, sensors, batteries and organic solar cells [20-23].

For practical application it was considered that efficiency of OSCs should be at least around of 5%. Calculation made by G. A. Chamberlain [24] showed that efficiency of about of 10% can be realized in Schottky barrier solar cells. Recently, the efficiency over 10% has been achieved in the organic photovoltaic cells [25, 26]. There are predictions that in the single junction organic solar cells more than 20% power conversion efficiency can be achieved [26, 27]. Different kinds of organic solar cells, including sing-layer, bi-layer or heterojunction and bulk heterojunction solar cells were fabricated and investigated [28-32]. Among the promising solar cells are cells having organic-inorganic heterojunction structures. For these cells polycrystalline or single crystal Si of n-type (or p-type) can be used which are several times cheaper with respect to p-n junction Si wafers [33]. One of the

fabricated organic-inorganic cells (Ag/n-GaAs/p-CuPc/Ag) showed efficiencies of around of 4% [34]. A tandem approach for organic semiconductor solar cells allows to improve performance of the devices due to overlapping of absorption spectra of the several cells [35, 36]. Key problem in practical utilization of the newly developed solar cells is cost of materials and fabrication technology. In this paper in continuation of our efforts in fabrication and investigation of solar cells [37], we are describing the properties of an organic-inorganic hybrid tandem bulk heterojunction cell based on AlPc:H<sub>2</sub>Pc bulk heterojunction and n-Si cells fabricated by developed by us pressing technology..

## 2. Experimental

The commercially available aluminum phthalocyanine (AlPc), metal free phthalocyanine (H2Pc), ITO coated plastic substrates and crystalline n-Si substrates were purchased from Acros and Aldrich company (AlPc and H<sub>2</sub>Pc) and Institute of Silicone Technology n-Si (doped by phosphorus of  $10^{18}$  cm<sup>-3</sup>), respectively. The molecular formulas of AlPc and H<sub>2</sub>Pc were C32 H16 Al Cl N8 and C32 H18 N8 respectively, while the orientation of silicon substrate was (100). Selection of particular AlPc and H<sub>2</sub>Pc organic semiconductors was done due to large differences of work functions: 3.8 eV (AlPc), 4.04 eV (H<sub>2</sub>Pc) that allow us to fabricate bulk heterojunction structure. Figure 1(a) and (b) show the molecular structure of aluminum phthalocyanine and metal free phthalocyanine, respectively. A mixture of 50 wt.% AlPc and 50wt.% H<sub>2</sub>Pc powders was used for deposition of the thin film by vacuum thermal evaporation.

A 170 nm thick film of AlPc:H<sub>2</sub>Pc blend was deposited onto the ITO coated plastic substrate and crystalline n-Si substrate by thermal evaporation. The thickness of the Si wafer was 0.5 mm and it was doped with phosphorus for solar cell applications. A 50 nm thick aluminum film was deposited on the back side of the n-Si substrate. The deposition process was also carried out by using AUTO 306 Vacuum thermal evaporator (Edwards) under a vacuum higher than  $10^{-6}$  Torr and the deposition rate was 0.1 nm/s. The thickness of the films was measured by FTM5 crystal oscillator, which works on micro-balancing technique for the simultaneous measurement of deposition rate and thickness of the film. The sizes of the AlPc-H<sub>2</sub>Pc films deposited on n-Si and ITO substrates were equal to 2.5:2.0 cm<sup>2</sup>. The samples of ITO and n-Si coated with AlPc-H<sub>2</sub>Pc were put face to face and pressed with a pressure of  $3.5310^{-1}$  to  $4.31 \ 10^{-1} \text{ N/cm}^2$ at 60-80 °C for 20-30 min. Figure2 shows the samples before (a) and after (b) pressing. The samples were fixed in pressed condition by using adhesive. The active area of

the fabricated sample was 3.0 cm<sup>2</sup>. Device architecture was the following:  $ITO/AIPc:H_2Pc/n-Si/AI$ .



*Fig.1. (a) molecular structures of aluminum phthalocyanine* (*AlPc*) and (b) metal free phthalocyanine (H<sub>2</sub>Pc)

Materials which were used for fabrication of the photoelectric cell have the following work functions: 4.7 eV (ITO), 4.1 eV (n-Si) and 4.3 eV (Al) [38-41]. It may be considered that AlPc and  $H_2Pc$  form the bulk heterojunction cell, n-Si forms the second cell where the top electrode is formed by AlPc:H<sub>2</sub>Pc film. Accordingly n-Si plays a role of bottom electrode of the AlPc:H<sub>2</sub>Pc bulk heterojunction photoelectric cell.

For measurement of voltage and current a digital multimeter HIOKI DT 4253 and for measurements of the temperature digital multimeter FLUKE 87 were used. As a light source a filament bulb was used and the intensity of the light was calibrated by using illuminometer Kyocera Corporation. Different illuminations were obtained by changing the distance between photoelectric cell and filament bulb.



Fig. 2. The ITO/AlPc:H<sub>2</sub>Pc/n-Si/Al photoelectric cell before (a) and after (b) pressing

# 3. Results and discussions

The surface morphology of AlPc-H<sub>2</sub>Pc film is characterized by atomic force microscopy (AFM). Figure 3 shows AFM images of the AlPc-H<sub>2</sub>Pc blend film. It is evident from Fig. 3 that the film is well developed and has sufficiently good roughness. This type of film is required for better harvesting of incident light. The optical characterization of the AlPc-H<sub>2</sub>Pc blend was carried out by UV-Visible spectroscopy. Fig. 4 shows absorption spectrum of the AlPc- $H_2Pc$  blend. The strong absorption at 715nm can be regarded to the transition from HOMO to the LUMO. The absorption spectrum has two wide ranges of high absorption in the visible spectrum range: from 350 nm to 400 nm, and from 550 nm to 800 nm.

Fig. 5 shows I-V characteristics of the ITO/AlPc:H<sub>2</sub>Pc/n-Si/Al tandem solar cell at dark condition and room temperature (T=25 °C). A forward bias potential was applied to the ITO. In Fig.5 the presence of a rectification behavior can be seen. A rectification ratio (RR), determined as  $I_F/I_R = 5.0$ , where  $I_F$  and  $I_R$  are forward and reverse bias currents at a voltage of 4 V. This kind I-V characteristics are very common in the case of organic semiconductors [17, 29]. Fig. 6 shows I-V characteristics of the tandem bulk heterojunction solar cell under illuminations of 290 W/m<sup>2</sup>, 240 W/m<sup>2</sup> and 200 W/m<sup>2</sup>. The obtained I-V characteristics have similarities with the characteristics of a number of organic semiconductors investigated before [17, 29]. One of the most important point of the I-V characteristics obtained under illumination condition (Fig.6) is the maximum power point (MPP). In [42] number of methods have been described for the determination of maximum power points. Moreover, the MPP tracking for modules is also described in [43]. The overview of MPP tracking control methods are presented in [44]. In this work we used the method which was developed in [45]. As an optimum voltage (V) in operating point 76% of the open circuit voltage ( $V_{oc}$ ) was selected and accordingly the density of currents (J) at maximum power point was found. For the three curves which are shown in Fig.6 the optimum voltage and the density of current were found accordingly for the illuminations of 290 W/m<sup>2</sup> (V<sub>1=</sub>0.243 V and J<sub>1</sub>=0.38 mA/cm<sup>2</sup>), 240 W/m<sup>2</sup>  $(V_{1=}0.241 \text{ V} \text{ and } J_{1}=0.29 \text{ mA/cm}^2)$  and 200 W/m<sup>2</sup>  $(V_{1=}0.239 \text{ V and } J_{1}=0.21 \text{ mA/cm}^2)$ . This method [49] is simple with respect of methods described in [42-44] and can be used for estimation of coordinates of maximum power point in I-V graphs.



Fig. 3. AFM images of the AlPc:H<sub>2</sub>Pc composite film



Fig. 4. Absorption spectra of a deposited blend of AlPc and  $H_2Pc$  film and  $J_{sc}$  of the ITO/AlPc: $H_2Pc/n$ -Si/Al photoelectric cell



Fig. 5. I-V characteristics of the ITO/AlPc:H<sub>2</sub>Pc/n-Si/Al tandem solar cell at dark condition



Fig. 6. I-V characteristics of the ITO/AlPc:H2Pc/n-Si/Al tandem solar cell under illuminations of 290 W/m<sup>2</sup>, 240 W/m<sup>2</sup> and 200 W/m<sup>2</sup>

One of the most important parameter of photoelectric cells is the stability. Investigation of the photoelectric properties showed that the organic semiconductor phthalocyanine based solar cell having the normal structure (ITO/PEDOT:PSS/Organic conventional laver/Al) degraded in 24 hrs after fabrication, whereas solar cells with an inverted structure (TO/TiO2/organic layer/PEDOT:PSS/Au) showed good stability in air compared to the cell with normal structure [46]. It was considered that degradation took place in the cells with normal structure due to oxidation of Al into Al<sub>2</sub>O<sub>3</sub> at the Al/organic film interface which decreases the conductivity of the initial metallic layer. Moreover diffusion of Al into the active layer of organic semiconductor may take place: Al in organic semiconductor plays the role of recombination sites that will decrease the efficiency of the cell [46]. On the other hand in the photoelectric cells with normal structure the corrosion of indium-tin-oxide (ITO) takes place due to the acidity of PEDOT:PSS (poly(3,4ethylenedioxylenethiophene):poly(4-styrene sulfonic acid) [5, 46]. Therefore, in the inverted structure Al is not used, and PEDOT:PSS is not contacted with ITO [46]. The efficiency of the inverted structure gallium phthalocyanine/PCBM (GaPc/PCBM) photoelectric cell was low (n= 0.059 %), but more or less stable. As seen from Fig. 2 for the tandem cell, the organic semiconductor film AlPc:H2Pc is in contact only with ITO and n-Si wafer, which may bring a longer life for this photoelectric cell. Long term stability of the cells will be a matter of future investigations. Previously we investigated the degradation of an organic-inorganic Ag/n-GaAs/p-CuPc/Ag photoelectric cell that was photoactive in UVvisible-IR wavelength region (200-1000 nm) [34]. A power conversion efficiency of the fresh cell was equal to 4% in the year 2000. In five years degradation in the efficiency was observed in an exponential behavior from 4% to 0.6%. After this time the efficiency of the cell was stabilized and the cell was tested and used as a photoelectric sensor as a teaching aid for ten years (up to 2015).

In this paper the presented data shows that in the fabrication of the ITO/AlPc:H2Pc/n-Si/Al photoelectric cell; firstly, a pressing technology is used to fabricate the photoelectric cells. Secondly, pressing of the film of the same composition AlPc:H<sub>2</sub>Pc made the process more reliable and properties of the cells more predictable. Thirdly, unlike many tandem and stacked solar cells [47, 48], the ITO/AlPc:H2Pc/n-Si/Al solar cells used no conductive layers between connecting the two photoelectric cells, as it was done in a heterojunction structure [39]. It means that the ITO/AlPc:H<sub>2</sub>Pc/n-Si/Al cell has structural features of a combined tandem and heterojunction photoelectric cell. Under illumination of 290  $W/m^2$  the values of V<sub>oc</sub>, I<sub>SC</sub>, FF and efficiency were equal to 0.32 V, 0.9 mA/cm<sup>2</sup>, 0.37 and 0.22 %, respectively. After one month of fabrication no visible changes in the photoelectric properties of the fabricated cell were observed. It is probably due to the adhesive (Fig.4b), which efficiently encapsulates the cell and isolate

it from the oxygen of the environment. It is a known effect that the annealing treatment, time, annealing temperature, and environmental conditions affect the properties of the organic solar cells [49]. The effect of annealing of the ITO/AlPc:H<sub>2</sub>Pc/n-Si/Al photoelectric cell and its optimization will be a matter of future work.

As it is known that absorption spectrum of Si covers wide range of wavelength (250-1350 nm) [50, 51] and definitely it will affect the spectrum of J<sub>sc</sub>. In Fig.4 absorption spectra of a deposited blend of AlPc and H<sub>2</sub>Pc film and the  $J_{sc}$  of the ITO/AlPc:H<sub>2</sub>Pc/n-Si/Al photoelectric cell are presented. It is seen that the absorption spectrum and spectrum of J<sub>sc</sub> have similarity and difference. The difference of the spectra probably is due to the contribution of Si-n. The Si-n is not "conductive electrode", it is semiconductor. Therefore under effect of light, which was not absorbed by AlPc:H<sub>2</sub>Pc film, photo induced charges are generated, concentration of total charges will increase that finally will affect the opencircuit voltage and short circuit current. More detailed investigations will be done in future in order to fully identify the role of bulk heterojunction film, optimize the thickness of the film, and finally increase of the efficiency of the cell. The current work was realized in continuation of our investigations of the properties of organic-oninorganic Ag/n-GaAs/p-CuPc/Ag photoelectric cell [34]. At the same time we would like to emphasize that the most important points are fabrication and investigation of the ITO/AlPc:H<sub>2</sub>Pc/n-Si/Al cell by using pressing technology which can decrease in principle the fabrication cost and finally the cost of energy which will be produced by the cell. Moreover, we also emphasize on the presence of two photosensitive structures (n-Si and AlPc:H<sub>2</sub>Pc) which are actually complementary by nature.

## 4. Conclusion

An organic-inorganic hybrid tandem bulk heterojunction ITO/AIPc:H<sub>2</sub>Pc/n-Si/Al photoelectric cell was fabricated by vacuum evaporation and pressing technology, I-V characteristics was investigated in dark conditions and under illumination. The fabrication technology of the cell can be used for the development of photoelectric cells technology, in particular, tandem cells. Moreover, the simple preparation procedure of the cells make them possible to use as a teaching aid.

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