

# Improvement of Solar PV Efficiency. Potential Materials for Organic Photovoltaic Cells

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**Abstract** – Organic photovoltaic (OPV) cells are considered as a viable alternative to those energy sources currently in use. In this work three derivatives of original N,N'-dimetilaminobenziliden-1,3-indandione (DMABI) material are presented as potential materials for OPV. The photoconductivity threshold energy was evaluated from the perspective of spectral dependence of photoconductivity quantum efficiency, and the optical energy gap was defined to determine the optical absorption spectra. The absorption spectra of derivatives are blue shifted compared to original DMABI. Use of these derivatives in multilayer solar cells with original DMABI makes it possible to broaden the spectral response range of OPV.

**Keywords** – organic photovoltaic cells, thin films, indandione, photoconductivity quantum efficiency, optical energy gap.

## I. INTRODUCTION

Renewable and clean energy is a challenge in present-day society. Therefore new energy sources are necessary to bring cheap and easy solutions into the market. Solar energy is a source that humans have used for ages. Only the last century has brought an opportunity to transform solar to electric energy using photovoltaic cells [1, 2]. The vast proportion of the photovoltaic market has filled up with Si based photovoltaic cells but the high production costs of such technology do not allow it to become a significant part of the energy market [3]. One solution to change this is the introduction of organic photovoltaic (OPV) cells. These cells offer lower costs, flexibility and easy, low-environmental impact technology [4-6]. Organic materials are applied increasingly for various electronics and OPV sector is one with the most potential [7-10]. Great steps have been taken to put them into practice in recent years; still more studies must be done to improve OPV parameters. The selection of materials is one of the options for OPV improvement as a wide range of organic materials can be used to broaden the spectral response range of OPV by using materials for which the absorption spectral region is different. This is one option for improving the efficiency of OPV cells [11, 12].

Knowledge on the energy level structure is essential in the material selection process. Charge carrier generation is influenced by an energy gap of material  $E_g$  which is common with the ionization potential of material in its solid state. This is an important parameter to create an energy level diagram for the solar cell [13, 14].

The energy of the photoconductivity threshold from the 'spectral dependence of photoconductivity quantum efficiency and the optical energy gap from the optical absorption spectra

were determined in order to understand the photoelectrical properties of chosen materials.

In this work N,N'-dimetilaminobenziliden-1,3-indandione (DMABI) derivatives of various functional groups were investigated. Functional groups were selected with the aim to improve energetic parameters and photoelectrical properties. The selection was also made with an emphasis on broadening the spectral response range of OPV. The maximum absorption capacities of the chosen derivatives are blue shifted compared to the DMABI and some other DMABI derivatives [15, 16].

## II. DESCRIPTION OF EXPERIMENTAL RESEARCH

### A. Materials

Three derivatives of DMABI were chosen for the study: DMABI-OH, MeOBI and MeSBI, where the N,N'-dimetilamino group is substituted by hydroxyl, methoxy and methylsulfanyl groups, respectively (see Fig.1).

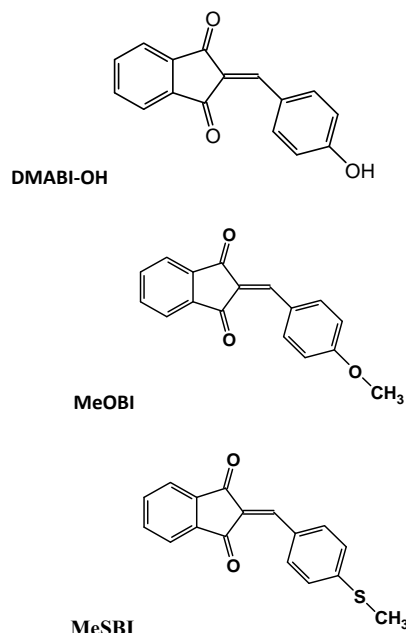


Fig. 1. Chemical structure of the compounds.

The DMABI molecule consists of an 1,3-indandione part as an electron acceptor and a dimetilaminobenzene part, as an electron donor. An exaction is formed by a photoinduced intramolecular electron transfer between the electron donor and the electron acceptor parts.

DMABI and its derivatives represent the class of photosensitive molecular crystals [17]. The previous research of DMABI thin films and host-guest systems demonstrates the perspective use of DMABI in OPV [18]. It has also been shown that the quantum efficiency of photoconductivity  $\beta(h\nu)$  for DMABI molecules in thin films is of the order of  $1 \cdot 10^{-2}$  el./phot. at 480 nm ( $\sim 2.6$  eV). The threshold energy of photoconductivity  $E_{th}$  has been estimated at  $E_{th} = 1.95 \pm 0.02$  eV [19, 20].

All the investigated derivatives were synthesized at the Institute of Applied Chemistry of the Riga Technical University [21].

### B. Preparation of samples

The typical sandwich-type structure for the samples was used where a single layer of organic material is sandwiched between two semi-transparent metal electrodes on a glass substrate. Nine identical active areas were made during evaporation – each at the point of intersection of the Au and Al electrode. The active area of the samples was  $4 \text{ mm}^2$  (see Fig. 2).

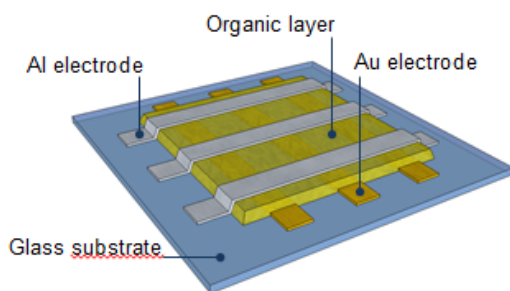


Fig. 2. Scheme of the sample

Two different metals were used for electrodes. Gold (Au) was used as the bottom electrode but, in case of the top electrode, Au diffusion into the organic layer was observed to take place after evaporation of Au due to a short circuit in the top electrode of the sample. Therefore aluminium (Al) was chosen for the top electrode since its evaporation temperature is much lower than Au and thereby it is better for protection of the organic layer.

The samples were fabricated by thermal evaporation in vacuum  $1 \cdot 10^{-6}$  mbar. The average thickness of the semi-transparent metal electrodes was 50 nm. The organic layers were sublimated at a temperature of  $170^\circ\text{C}$  with a thicknesses of: 320 nm for DMABI-OH, 620 nm for MeOBI and 530 nm for MeSBI (the thickness was measured by the surface profilometer Veeco Dectac-150 after evaporation). As the layer surface roughness of the organic materials was too high to place the top electrode successfully, the substrate was cooled to the temperature of  $-100^\circ\text{C}$ . Thus it was possible to considerably reduce the roughness of the organic layer and facilitate formation of a smooth aluminium layer on top of it. All layers were made without breaking the vacuum between evaporation steps. The samples for which organic molecules were mixed with poly(methyl methacrylate) (PMMA) were made to obtain the absorption spectra for the determination of

an optical band gap as it was not possible to do so for the evaporated thin films due to high roughness of surface that caused high light scattering in the layer.

Therefore thin films of DMABI derivatives mixed with PMMA (5% organic material + 95% PMMA mixed in solvent consisting of 5:1 acetone + chloroform) were spin-coated on quartz substrates at 800 rpm.

The absorption spectra of organic layers and transmission spectra of metal layers were measured by UV-NIR spectrometer Ocean Optics HR400 in order to determine the optical energy gap and calculate the photoconductivity quantum efficiency.

### C. Photoelectrical measurements

The samples were illuminated by xenon lamp TYP 1450 through interference filters for photoconductivity spectrum measurements. The spectral range between 700 nm and 300 nm with a step 10 nm was selected. The photocurrent measurements were carried out at both polarities by illuminating both Au and Al electrodes. The electrometer Keithley 651 with an incorporated DC voltage source was used for electrical measurements.

Current-voltage characteristics were measured without illumination to see the quality of the sample and to define the voltage value where the character of the current-voltage curve is ohmic. Under these conditions, charge carrier injection from the electrodes has not yet started and the conductivity of the sample has been provided only by charge carriers in thermal equilibrium. According to the current-voltage characteristics, the voltage of 10V for MeOBI and 2V for DMABI-OH and MeSBI were chosen for photoelectrical measurements.

The photoelectrical measurements were performed in a cryostat with a vacuum  $\sim 1 \cdot 10^{-5}$  mbar.

## III. RESULTS AND DISCUSSION

A number of methods may be applied to characterize the energy gap of organic molecules in a solid state. Charge carriers are not quasi-free electrons and holes in case of organic crystals. They form a polaron-type quasi-particle enveloped in electronic and vibronic polarization clouds. The energy difference between the electronically relaxed hole and the electron is attributed to an optical band gap  $E_G^{Opt}$  [14, 22]. The low energy threshold of the organic thin film absorption spectra may be used to obtain  $E_G^{Opt}$ .

The electronically and vibrationally relaxed state difference of holes and electrons corresponds to the adiabatic energy gap  $E_G^{Ad}$ . The value of the adiabatic energy gap  $E_G^{Ad}$  correlates with the value of the threshold energy of photoconductivity  $E_{Th}$ ,  $E_G^{Ad} = E_{Th} + \Delta E$ , where  $\Delta E$  is effective relaxation energy of ion pair state [14, 22].

The quantum efficiency of photoconductivity  $\beta(h\nu)$  makes it possible to estimate the threshold energy of photoconductivity  $E_{th}$ . The values of the quantum efficiency of photoconductivity were obtained by the equation:

$$\beta(h\nu, U) = \frac{j_{ph}(h\nu, U)}{k(h\nu)I(h\nu)g(h\nu)}, \quad (1)$$

Where:

$j_{ph}$  - density of the photocurrent at a given photon energy  $h\nu$  and applied voltage  $U$ ;

$I(h\nu)$  - intensity of light ( $\text{phot}/\text{cm}^2\text{s}$ );

$k(h\nu)$  - transmission of semi-transparent electrode;

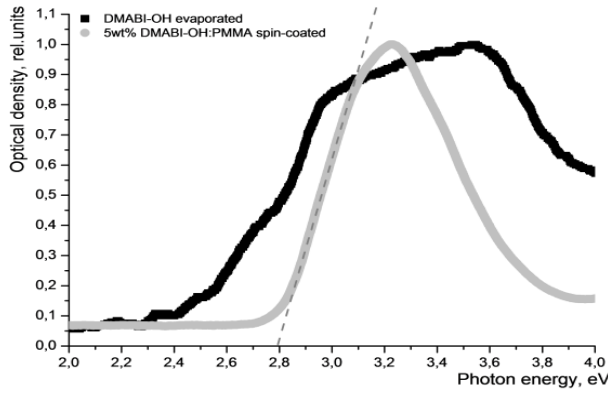
$g(h\nu)$  - the coefficient which characterises absorbed light in organic layer  $g(h\nu)$  can be obtained by the equation:

$$g(h\nu) = \int_0^L \exp[-\alpha(h\nu)x]dx = 1 - \exp[-\alpha(h\nu)L], \quad (2)$$

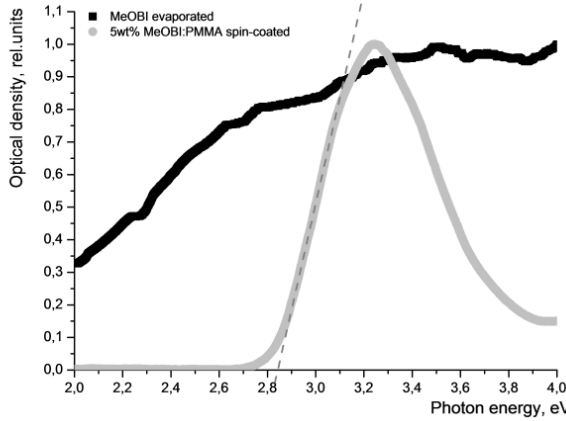
Where:

$\alpha(h\nu)$  - the absorption coefficient

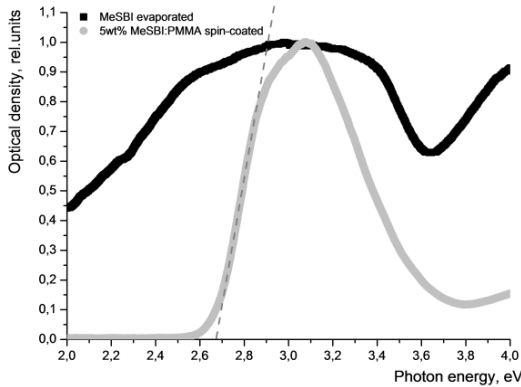
$L$  - the thickness of a sample [14].



A



B



C

Fig. 3. Absorption spectra of DMABI-OH (a), MeOBI (b), MeSBI (c) evaporated and spin-coated samples (optical band gap is determined from absorption spectra for spin-coated samples).

The previous researches demonstrate that in anthracene-type crystals the spectral dependence of  $\beta(h\nu)$  in the near threshold region can be approximated by:

$$\beta(h\nu) = A(h\nu - E_{th})^n, \quad (3)$$

where  $n \approx 5/2$  and  $E_{th}$  is the threshold energy of photoconductivity,  $A$  is coefficient [14, 22].

The absorption spectra were obtained for both evaporated and spin-coated samples (see Fig. 3 a,b,c).

The absorption spectra for evaporated films were obtained broadly and the maximum was difficult to determine. It is not clear whether this is an effect of the material. One of the reasons for this could be high light scattering due to roughness of the surface of evaporated films as in case of the evaporation of polycrystalline structures are formed. Therefore compounds were mixed in PMMA matrix. The absorption maximum for all three compounds obtained from the PMMA matrix was within 3.1 to 3.3 eV. It is shifted to the ultraviolet region compared to the original DMABI which holds the absorption maximum of 2.583 eV [23].

The absorption spectra of sublimed samples were not possible to use for optical band gap determination either. Therefore these values were also obtained for samples consisting of PMMA (see Fig. 3). The highest value was obtained for MeOBI,  $E_G^{Opt} = 2.85 \pm 0.03 \text{ eV}$ . The optical band gap values for DMABI-OH and MeSBI were  $E_G^{Opt} = 2.80 \pm 0.03 \text{ eV}$  and  $E_G^{Opt} = 2.68 \pm 0.03 \text{ eV}$ , respectively (see Table 1). As mentioned previously, broadening the spectral response range of OPV by using materials for which the absorption spectral region is different is one of the options on how the efficiency of the OPV cell can be improved.

In figure 4 the comparison of the absorption spectra is given. The blue line corresponds to the absorption maximum for the original DMABI [23]. This makes it possible to use these in tandem solar cells as the compounds may absorb ultraviolet light and let through the red light absorbed by, for example, silicon. Thus, by combining several materials with different absorption, it is possible to use a greater part of sun light.

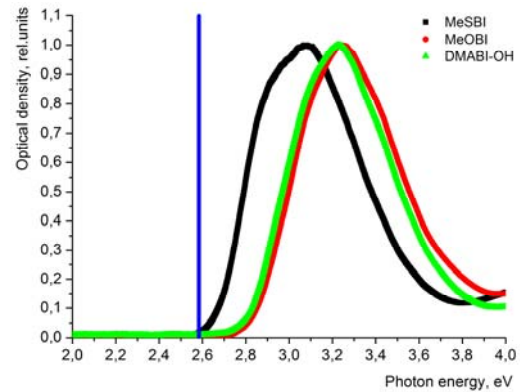


Fig. 4. Comparison of absorption spectra of investigated compounds with DMABI (spin-coated samples). The blue line corresponds to the absorption maximum of DMABI

The quantum efficiency of photoconductivity  $\beta(h\nu)$  was calculated according to expression (1). The spectral dependencies of  $\beta(h\nu)$  on illumination through Au and Al electrodes of both polarities were compared (see Fig. 5-7).

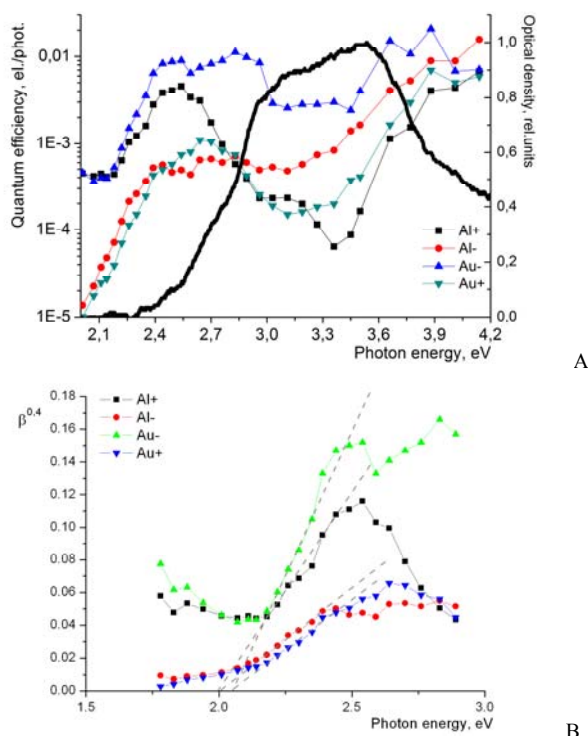


Fig. 5. Spectral dependences of quantum efficiency of photoconductivity  $\beta(h\nu)$  of Au/DMABI-OH/Al and absorption spectra of evaporated DMABI-OH (a); determination of the values of threshold energy of photoconductivity  $E_{th}$  (b). (Al+ (illuminated through Al electrode, voltage positive), Al- (illuminated through Al electrode, voltage negative), Au- (illuminated through Au electrode, voltage negative), Au+ (illuminated through Au electrode, voltage positive))

The character of the  $\beta(h\nu)$  spectra for the derivatives was relatively independent of electrode polarity and the illuminated electrode. If compared to the absorption spectra, for DMABI-OH two maxima at long and short wavelengths sides of the absorption maximum were observed. The anti-correlation between  $\beta(h\nu)$  and absorption curves corresponds to the case that the photogeneration process takes place in the volume instead of interface between electrode and organic film. In case of DMABI-OH the values of  $\beta(h\nu)$  for negative Au electrode were about one order higher than for positive Au. This observation could have resulted from the fact that the influence of charge carrier transport as  $\beta(h\nu)$  is affected not only by photogeneration, but also by transport of charge carriers. In case of MeOBI and MeSBI, the difference between the character of  $\beta(h\nu)$  spectra of electrode polarity and illuminated electrode was not explicit (see Fig. 6a, 7a). That could point to equal transport of electrons and holes in this compound. It is good property of material for OPV as a majority of materials used for OPV are either electron transport materials, or hole transport materials.

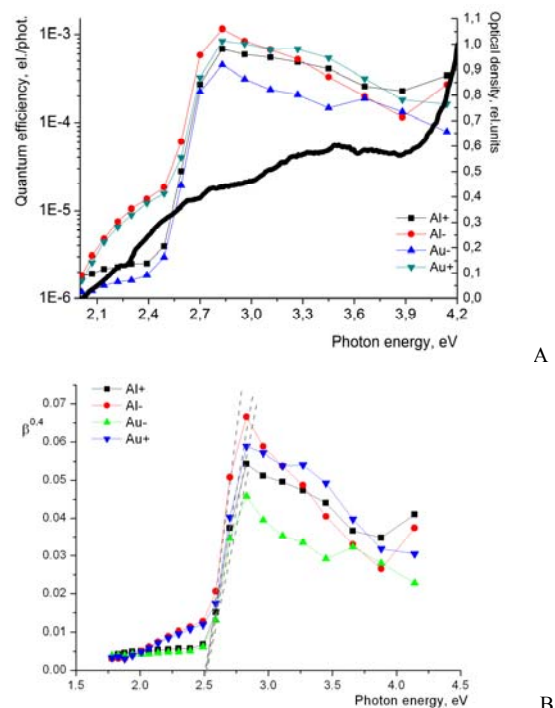


Fig. 6. Spectral dependences of quantum efficiency of photoconductivity  $\beta(h\nu)$  of Au/MeOBI/Al and absorption spectra of evaporated MeOBI (a); determination of the values of threshold energy of photoconductivity  $E_{th}$  (b). (Al+ (illuminated through Al electrode, voltage positive), Al- (illuminated through Al electrode, voltage negative), Au- (illuminated through Au electrode, voltage negative), Au+ (illuminated through Au electrode, voltage positive))

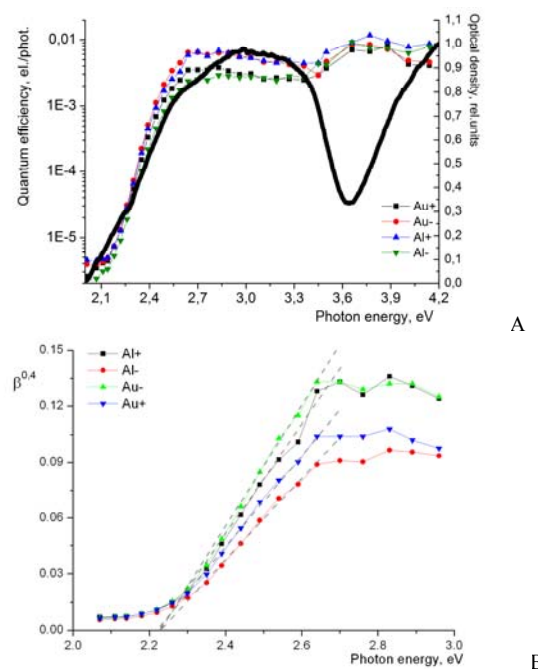


Fig. 7. Spectral dependences of quantum efficiency of photoconductivity  $\beta(h\nu)$  of Au/MeSBI/Al and absorption spectra of evaporated MeSBI (a); determination of the values of threshold energy of photoconductivity  $E_{th}$  (b). (Al+ (illuminated through Al electrode, voltage positive), Al- (illuminated through Al electrode, voltage negative), Au- (illuminated through Au electrode, voltage negative), Au+ (illuminated through Au electrode, voltage positive))



The highest values of  $\beta(h\nu)$  were observed for DMABI-OH and MeSBI and reached up to  $1 \cdot 10^{-2} \text{el./phot.}$  at plateau region of photon energy 2.4 to 3.0 eV for DMABI-OH and at 3.8 eV for MeSBI (see Fig. 5a). In case of MeSBI, it is the maximum point but values of  $\beta(h\nu)$  are close to  $1 \cdot 10^{-2} \text{el./phot.}$  at all plateau regions 2.5 to 4.0 eV (see Fig. 7a). These values repeat the previous high results of DMABI [20, 21].

The lowest  $\beta(h\nu)$  maximum value was obtained for MeOBI and reached  $1.15 \cdot 10^{-3} \text{el./phot.}$  at 2.8 eV (440 nm) (see Fig. 5a).

The values of threshold energy of photoconductivity  $E_{th}$  were determined according to the expression (3). The threshold energy of photoconductivity for DMABI-OH was obtained  $E_{th}=2.05 \pm 0.03 \text{ eV}$  (see Fig. 5b), for MeOBI  $E_{th}=2.44 \pm 0.03 \text{ eV}$  (see Fig. 6b) and the value of threshold energy of photoconductivity for derivative MeSBI  $E_{th}=2.23 \pm 0.03 \text{ eV}$  (see Fig. 7b). All experimentally obtained values are summarized in Table 1.

TABLE I

THE THRESHOLD ENERGY OF PHOTOCONDUCTIVITY  $E_{th}$  AND OPTICAL ENERGY GAP  $E_G^{Opt}$  VALUES OF DMABI-OH, ME OBI AND MESBI.

Compound	$E_{th}$ (eV)	$E_G^{Opt}$ (eV)
DMABI-OH	$2.05 \pm 0.03$	$2.80 \pm 0.03$
MeOBI	$2.44 \pm 0.03$	$2.85 \pm 0.03$
MeSBI	$2.23 \pm 0.03$	$2.68 \pm 0.03$

Determination of the energy gap of organic materials is also a significant part of solar cell fabrication. When using more than one material it is necessary to know their compatibility by making energetic diagrams. The determined values provide information on the investigated compound's place in the energetic diagram.

TABLE II

INCREASE OF THRESHOLD ENERGY OF PHOTOCONDUCTIVITY  $E_{th}$  AND OPTICAL ENERGY GAP  $E_G^{Opt}$  VALUES OF DMABI-OH, ME OBI AND MESBI.

Compound	$E_{th}$ (eV)	$E_G^{Opt}$ (eV)
DMABI-OH	$0.10 \pm 0.03$	$0.65 \pm 0.03$
MeOBI	$0.49 \pm 0.03$	$0.70 \pm 0.03$
MeSBI	$0.28 \pm 0.03$	$0.53 \pm 0.03$

In both cases the  $E_{th}$  and  $E_G^{Opt}$  values are greater for derivatives than for original DMABI ( $E_{th}=1.95 \pm 0.02$  and  $E_G^{Opt}=2.15 \pm 0.03$  [20, 21, 24]), (see Fig. 8). The benefit of substitution of functional groups is summarized in Table 2.

Increase of the energy gap means that these compounds will absorb light close to the ultraviolet region but the width of the absorption spectrum for them will be narrower. This allows the use of compounds for tandem solar cells in combination with other compounds.

Increase of the energy gap is related to efficiency of solar cell. One of the characters that influence efficiency is open circuit voltage. The energy gap of material is the difference between the highest occupied molecular orbital level (HOMO) and the lowest unoccupied molecular level (LUMO). In a two-material system, open circuit voltage is mainly determined by the difference between the HOMO level of donor material and

LUMO level of acceptor material [25, 26]. Use of the investigated materials in combination with materials with similar HOMO and LUMO levels and a wide energy gap would increase the open circuit voltage.

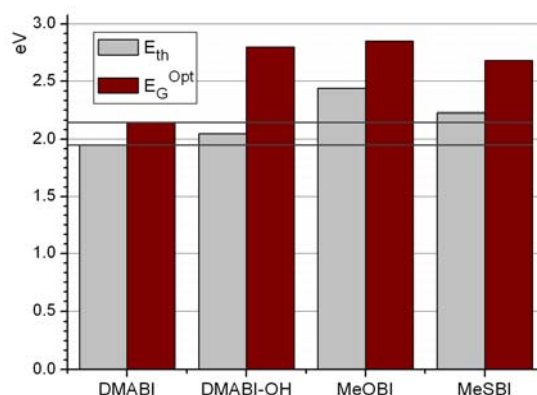


Fig. 8. Threshold energy of photoconductivity  $E_{th}$  and optical energy gap  $E_G^{Opt}$  values of DMABI-OH, MeOBI and MeSBI molecules compared with values of DMABI [20, 21, 24].

#### IV. CONCLUSIONS

Investigation of new materials gives the possibility to find materials with better properties and, in combination with several materials increase the efficiency of solar cells.

In comparison to DMABI, the studied compounds have a wider energy gap - higher energetic threshold where absorption starts. This property could be used by making tandem solar cells. The shifting of the absorption spectra of investigated compounds gives the possibility to broaden the spectral response range of OPV cells in combination with materials with different absorption. These compounds could absorb light in blue-ultraviolet region while passing through, for example, red light absorbed by another layer. A wider energy gap also influences the efficiency of a solar cell by increasing open circuit voltage.

The experiments showed that the acquisition of absorption spectra of compounds that tend to make polycrystalline thin films could be difficult because of their polycrystalline structure. To avoid that, it was necessary to mix these compounds in the PMMA matrix.

High roughness of surface of films points to the fact that the investigated compounds could be used in polymer OPV cells. Films in mixture with polymer are smooth and this eases the production and reduces the costs of OPV cells as the substrate does not need to be cooled. Investigated compounds could also be used for evaporated OPV cells as a lower layer covered by some other organic material which makes the surface of the film smooth. In that case, the compounds could increase the efficiency of OPV cells not only by broadening the spectral response range of OPV cells, but also by increasing the interface area of two materials. The surface roughness of the lower layer also stimulates light scattering. This permits the absorption of more light, thereby increasing the efficiency of the solar cell.

## ACKNOWLEDGMENTS

This work has been supported by the European Social Fund within the project «Support for Doctoral Studies at University of Latvia».

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