

SLOVAK UNIVERSITY OF TECHNOLOGY IN BRATISLAVA FACULTY OF ELECTRICAL ENGINEERING AND INFORMATION TECHNOLOGY

Ing. Peter Juhász

Author's report on the dissertation thesis

STUDY OF CHARGE TRANSPORT IN ORGANIC SEMICONDUCTORS BY ELECTRICAL TRANSIENT METHODS

for the acquisition of: academic title philosophiae doctor, PhD

in PhD. study programme: in študijnom odbore

Electronics and Photonics

5.2.13 Electronics

Date and place: In Bratislava, July 2016



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1 Introduction

The most electronics devices are based on the classical materials such as silicon (Si), galliumarsenide (GaAs) and other well-known inorganic materials. Nowadays, electronic devices based on these material already reached very high level and further improvement is difficult. Therefore, the research has been focused on development of new materials, which can provide alternative and/or replacement of the inorganic semiconductors. Recently, various materials have been proposed such as carbon nanotubes, graphene, molybdenum disulfide, or organic semiconductors. The organic semiconductors have been envisioned as materials that combine the electronic advantages of inorganic semiconducting materials with the optical, chemical, and mechanical benefits of organic compounds such as plastic materials.

Nobody thought that is the right way until the three scientists Alan J. Heeger, Alan G. McDiarmid, and Hideki Shirakawa in 1970's observed the electrical conductivity of conjugated molecules. Their invention was honored by the Nobel prize in Chemistry in 2000. After this success the organic electronics has witnessed enormous world-wide effort both in basic scientific research as well as in industrial development within the last decades.

As a result, in the last decades' new electronic devices such as organic field-effect transistor (OFET), organic light-emitting diodes (OLED) and organic solar cells have arrived and attract the attention of scientific and industrial research. These electronic devices have excellent and unique features such as mechanical flexibility, low-cost fabrication, optical transparency, etc. which find the application in displays of mobile phones, flexible solar cells and so on. Although there are some commercial application available on the market, the deep understanding of film growth and structure, electrical properties, and interface phenomena is still needed.

On a world-wide scale the research in this field can be divided on three main areas: (*i*) the design, technology, and fabrication of device, (*ii*) characterization of surfaces, interfaces, and thin films and, (*iii*) properties at molecular level.

The dissertation thesis deals with organic electronics devices, characterization of the electronic properties of structures based on thin films, and discussion on the origin at nanoscale and molecular level. The study of electrical properties is performed by steady-state current-voltage measurements, impedance spectroscopy (IS) and transient capacitance measurements; however, to reach the complex characterization of thin films this thesis needs to cover all three above-mentioned. The thesis is focused on the design of methodology suitable for analysis of charge carrier injection and charge carrier transport in organic semiconductors. Both frequency- and time-domain transient methods are compared with the steady-state approach. Common approaches in measurement analysis is not suitable since organic semiconductors wide band gap materials or insulator rather than semiconductors. It is also well-known that these materials have a greatly larger relaxation time than inorganic materials, that makes a challenge for application of these characterization techniques to this specific kind of materials.

All executed experiments such as device design, fabrication, and characterization of injection and transport properties took place at the Slovak University of Technology, Faculty of Electrical Engineering and Information technology, Institute of Electronics and Photonics.

1.1 Thesis of dissertation work

The organic devices are the center of world-wide application and research field. The thin film characterization is one of the employment area of research. Following goals of the dissertation thesis have been established:

- Perform analysis of data provided by the steady-state current-voltage (J-V), impedance spectroscopy (IS), and transient capacitance techniques of the metal-organic semiconductormetal structures, where the semiconductor material is pentacene. Finding a correlation between these methods and explanation of differences.
- 2. Perform the voltage dependence study of the relaxation processes activation energies on abovementioned devices.
- 3. Investigate the selected organic materials (pentacene, poly(3,4-ethylenedioxythiophene)poly(styrenesulfonate)) with other transport processes (hopping or band transport)
- Application of developed approach to the selected structure such as organic light-emitting diodes (OLED).

2 **Experiments**

Studies of charge injection and transport became of prominent importance to provide deep understanding of the charge relaxation processes in organic semiconductors. The scientific research carried out on charge transport mobility constitutes a key role for further optimization of light emission in OLEDs. Furthermore, the performance of organic electronics devices depends strongly on the quality of the semiconductor which is greatly affected by the defect states. The formation of defects in organic materials is still not well understood, and consequently, it is difficult to control the defect states in organic devices. Furthermore, it is a great challenge to distinguish the charge traps from the other phenomenon such as interface charging, injection barrier, or temperature dependence of the mobility.

This thesis is composed as a collection of original research papers related to the study of charge transport in organic semiconductors. Major part of the results has been published in international peer-review journals recognized in Master Journal List (the Current Content database) of the Thomson Reuters

corporation, some of the preliminary results have been presented on international conferences. The studies can divide as following:

- 1. Finding the appropriate measured methods and application these methods to organic semiconductors. Estimation of the correlation between these methods;
- Investigation of the voltage dependences of the activation energies of relaxation processes in organic diode;
- 3. Identification of different transport processes in selected organic solids (small molecules and polymers);
- 4. Application of developed approaches to the OLED devices.

The following sections explain the original contributions reported in four published journal papers and one conference contribution and their relation to the assignment and goals of the thesis. The research papers are denoted as P1-P5.

2.1 Correlation between steady-state current-voltage, impedance spectroscopy, and transient capacitance techniques

Various electrical and optical methods have been proposed for characterization of defects states and relaxation processes in inorganic semiconductors. The application of these methods to characterize the organic semiconductors is still a challenge and the deep analysis is required. For example, one of limiting factors is that organic semiconductor has a longer relaxation time than inorganic. As a result, the dielectric relaxations is much more difficult to distinguish from the trap-related processes. Therefore, for better characterization and in order to get more information about the defect states and relaxation processes, a combination of different measurement systems is required. This section describes the correlation between the steady-state method and two independent transient methods. The steady-state method is the current-voltage measurement. The two transient methods are represented by the impedance spectroscopy and the transient capacitance measurement.

Publication P1: Characterization of charge traps in pentacene diodes by electrical methods

The correlation between these measurement methods are reported in research paper published in the *Organic Electronics* journal. These experiments were done on pentacene as the well-known organic semiconductor. Pentacene (Fig. 1.) is mainly used for fabrication of the organic field-effect transistors (OFET) as a material with *p*-type conductivity. Pentacene is classified as a small molecule organic material and belongs to polycyclic aromatic hydrocarbons. Since HOMO and LUMO energies for pentacene are 5.0 and 3.2 eV, respectively, the metals Au (5.3 eV) and Al (4.2 eV) have been chosen as the electrode materials as the typical representatives of low and high injection metals for pentacene, respectively, to fabricate diode structure.

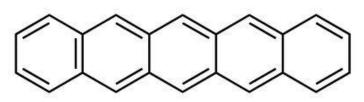


Fig. 1. Pentacene molecule

The diode has been characterized in temperature range 150K to 400K by steady-state current-voltage, impedance spectroscopy, and transient capacitance measurements.

From the analysis of the measured spectra the following statements are concluded:

- 1. The steady-state current-voltage measurement revealed the main bottleneck of charge transport.
- 2. The impedance spectroscopy measurements *distinguished the major charge relaxation to two processes*.
- 3. The transient capacitance measurement distinguished *charge relaxations even with very similar activation energies*.

Tab. 1. Summary of the activation energies evaluated by different characterization techniques.

Technique	ΔE_{T1} (eV)	ΔE_{T2} (eV)	ΔE_{T3} (eV)	ΔE_{T4} (eV)	ΔE_{T5} (eV)	ΔE_{T6} (eV)
Steady-state current-voltage Impedance spectroscopy Transient capacitance spectroscopy	0.29–0.34 0.27 0.25	0.26	0.31 0.31	0.32	0.43	0.46

Publication P2: Defect Analysis of Pentacene Diode

This paper was published in *Acta Physica Polonica A* journal and it aims on the impact of effective masses on data evaluation. It has been mentioned above that the localized states between HOMO and LUMO are assigned to the charge traps. The traps can be described with the three main parameters: the activation energy, which describe where this defects are places in energy diagram, the capture cross-section which defines the narrow of the traps and the trap density. These three parameters have obtained from the transient capacitance measurements using the DLTS evaluation approach. While the activation energy (2.2) is directly extracted from the Arrhenius curve (2.1)

$$ln\left(\frac{e_{\rm n}}{T^2}\right) = \ln(\gamma_{\rm n} \cdot \sigma_{\rm n}) - \frac{\Delta E}{k \cdot T}$$
(2.1)

$$\Delta E_{\rm T} = -k \cdot A_{\rm R} \tag{2.2}$$

where γ_n is semiconductor material parameter σ_n is capture cross section. The capture cross-section (2.3) depends on the effective mass of free charge carriers.

$$\sigma_{\rm n} = \frac{1}{\gamma_{\rm n}} expB_{\rm R} \tag{2.3}$$

The trap concentration depends on the amount of charge of doping materials, but this parameter cannot evaluate for the organic semiconductors because they are no doped. These materials using the conductivity of injected charges from the electrodes.

For the most of organic materials the effective mass is unknown, but for the pentacene is known three values: $1.3m_0$, $3.31m_0$, and $5.52m_0$ ($m_0=9.11 \times 10^{-31}$ kg). In this paper we investigated these three values of effective mass impact to determination the capture cross section parameters of traps. Our analysis pointed out that the higher value of effective mass leads to the lower effective capture cross-section. These values of effective mass would not result big differences in the capture cross-section values. The resulted value of effective capture cross-section is in one order of magnitude.

Tab. 2. The capture cross-sections for three values of hole effective masses

Deep		Capture cross-section $\sigma_{\rm T}$ [cm ²]				
level	energy $E_{\rm T}$ [eV]	$m^*/m_0 = 1.3$	$m^*/m_0 = 3.31$	$m^*/m_0 = 5.52$		
T1			3.7×10^{-18}	2.08×10^{-18}		
T2		3.56×10^{-18}		1.09×10^{-17}		
T3	0.63	5.16×10^{-15}	2.9×10^{-15}	1.29×10^{-15}		

2.2 The voltage dependences of the activation energies of relaxation processes in organic diode

The charge relaxation phenomenon is a process of transition to the new steady state after perturbation. This transition is a complex process depending on many parameters such as temperature or local electric field. Hence, the parameter describing the energy difference between these states, the activation energy, is not only simple constant, but it can be a function of the applied voltage. This approach has its own advantages as well as disadvantages. It is a great assistance for the estimation of the origin of the charge relaxation process since the electric field dependence may reveal the origin of relaxation. On the other hand, additional independent variable makes all measurements very time-consuming. Although finding suitable measurement parameters to fit measurement ranges costs certain time, the time required for evaluation processes significantly rises in geometric series.

Publication P1: Characterization of charge traps in pentacene diodes by electrical methods

The research paper among the finding the correlation between the electrical transient methods shows the voltage dependence of activation energy (Fig. 2. (c) shows the voltage dependency of activation energy of conductivity estimated from the steady-state current-voltage dependencies).

From the comparison of this dependence with the steady-state current-voltage characteristics it can be concluded follows: at the low voltages (<1 V) the current linearly depends on the applied voltage which corresponds with the Ohmic behaviour. Over the 1 V the current follows power dependence on the applied voltage slope and the power increased dramatically which is in good agreement with the increasing the activation energy. This increasing reaching the maximum around 2 V and this region called the trap-fill limit (TFL), where all localized levels are filling with injected charge carriers. Over this voltage the current voltage characteristics follow power law with power approaching the SCLC model with traps and activation energy is slowly decreasing too. This gradual decreasing of the activation energy is caused by continuous filling of the localized states. As a result, injected charge carriers are free to move from one electrode to the second electrode and this region is exhibiting behavior of space-charge limited current with lower concentration of trap states.

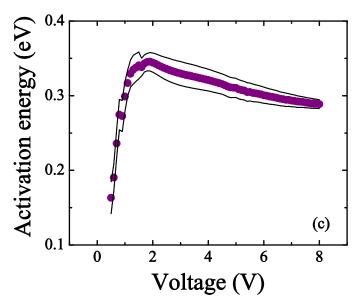


Fig. 2. Voltage dependency of activation energy of the conductivity

Publication P3: Charge injection and transport properties of an organic light-emitting diode (ADC 02)

This research paper shows the voltage dependence of activation energy estimated from the currentvoltage and impedance spectroscopy measurements in Fig. 3. The conductivity activation energy estimated from the steady-state measurements again shows the increasing of value by the dramatically increasing of current density in the voltage range of TFL (2~3V). After this increasing the activation energy voltage dependencies slightly decreasing. In contrast to the steady-state analysis, the impedance spectroscopy analysis gives information on two relaxation processes at low voltage region. One stands for the filling of the interfacial states and slightly increasing during the filling of unoccupied states. The other one is constant and represents the interface between indium tin oxide (ITO) and organic semiconductor layer (α -NPD). At the higher voltages the two relaxation are combined to one. The resulting relaxation activation energy is in great agreement with the activation energy extracted from the steady-state measurements. The charge relaxation represents the charge injection over the energy barrier at the ITO/ α -NPD interface. The applied voltage consequently suppresses the energy barrier and the charges are injected as described by the thermionic injection model.

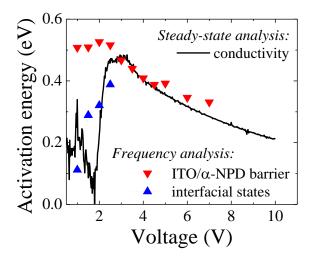


Fig. 3. Evaluated voltage dependence of activation energy

2.3 Identification of different transport processes in selected organic solids (small molecules and polymers)

The charge transport processes in organic materials can be devided to two cathegory *band transport* and *hopping transport*. The small molecule materials, like pentacene usually exhibit the band transport properties, while polymers, like PEDOT:PSS transport charge carriers by hopping process. The transport processes should be distinguishable from the temperature dependences of electrical conductivity.

Publication P1: Characterization of charge traps in pentacene diodes by electrical methods (ADC01)

This research paper is focused on the correlation between electrical characterization methods and the voltage dependence of activation energy. However, the analysis of the electrical conductivity revealed simple exponential dependence on the device temperature. In other words, the small-molecule-based device exhibited the band transport which can be described by following relation,

$$\sigma = \sigma_0 exp\left(-\frac{\Delta E}{k_B T}\right) \tag{2.4}$$

The Arrhenius plot verified linear relation between logarithm of electrical conductivity and the reciprocal temperature (Fig. 4).

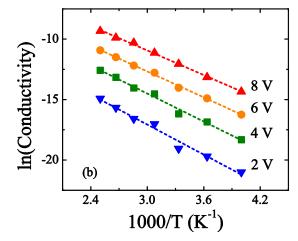


Fig. 4. Arrhenius plot of conductivity for selected voltages.

Publication P4: Secondary Doping in Poly(3, 4-ethylenedioxythiophene): Poly(4styrenesulfonate) Thin Films

This research paper deals with the electrical and structural properties of poly(3,4 – ethylenedioxythiophene):poly(4-styrenesulfonate) (PEDOT:PSS) thin films and different concentrations of selected secondary dopants. This organic material has big potential to replace transparent conductive oxide layers such as indium tin oxide or doped zinc oxide in organic solar cells, organic light emitting diode, or touch screens. The molecular structure of this material shown in Fig. 5, it is a mixture of two polymers, PEDOT and PSS. We have demonstrated that the secondary dopants are not present in the fabricated PEDOT:PSS films, but they greatly increase the conductivity by three orders of magnitude and influence the film structure.

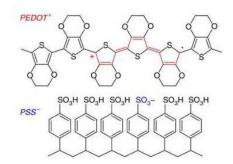


Fig. 5. Chemical structure of PEDOT:PSS

The PEDOT:PSS represent a strongly disordered polymer chains with localized charge-carrier states. The electrical conductivity of such systems can be described by the Motts variable-range hopping (VRH) model:

$$\sigma \propto \sigma_0 exp\left(-\left(\frac{T_0}{T}\right)^{\alpha}\right) \tag{2.5}$$

where the T_0 is the characteristic temperature representing the effective barrier to hopping of charge carriers between localized states, while the power coefficient α is equal

 $\alpha = 1/(1 + D)$, where *D* represent the dimensionality of the system. The analysis of the power coefficient α has been carried out by the evaluation of the maximum value of the correlation coefficient.

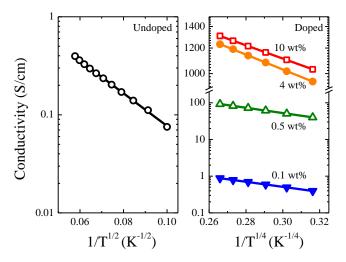


Fig. 6. Temperature dependences for undoped PEDOT:PSS and PEDOT:PSS doped by DMSO of various concentration

In the case of pristine PEDOT:PSS layer the power coefficient α is equal to $\frac{1}{2}$. This value represents the 1D charge transport. The power coefficient of doped PEDOT:PSS films is equal to $\frac{1}{4}$ this value describe the 3D charge hopping transport. These results are showed in the Fig. 6. In addition the continuous transition from quasi-1D VRH to 3D VRH (Fig. 7) has been demonstrated by gradual increasing of the dimethyl sulfoxide (DMSO) dopant concentration.

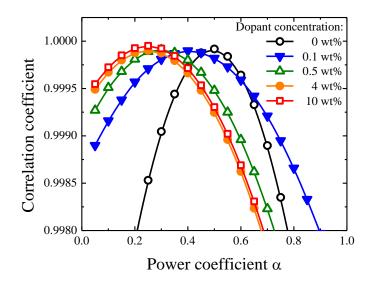


Fig. 7. The analysis of the power exponent α

2.4 Application of developed approaches to the OLED devices

After successful application of measurement methodology to simple metal-organic semiconductormetal devices and detail understanding to the working principles of devices the research has been focused on analysis of more complex device structures with two different organic semiconductors, namely the organic light-emitting diode (OLED).

Publication P3: Charge injection and transport properties of an organic light-emitting diode

This research paper is focused on the charge behavior study by the simple steady-state current-voltage and impedance measurements of the double-layer OLED device. The current-voltage measurements revealed that at higher voltages the measured current-voltage characteristics pointed out the double-carrier space-charge limited current, which can be describe by the following equations:

(2.6)

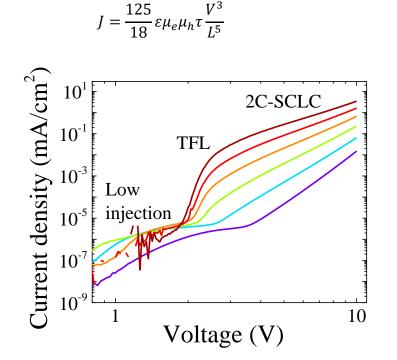


Fig. 8. Steady-state current-voltage characteristics in log-log scale illustrate two-carrier space-charge limited condictions

The impedance spectroscopy measurement at the same time pointed out that at low voltages there are two relaxation processes from which one relaxation activation energy is increasing by the voltage to the 3V. The second relaxation activation energy is constant 0.5 eV until 3 V than decreasing by increasing the voltages. The decay of activation energy follows the square root of applied voltage which can be assigned to the Pool-Frenkel model or thermionic emission.

As a result, the energy was compared with the energy diagram and the relaxation has been assigned to an injection of the charge over the energy barrier lowered by the external field. Besides the first relaxation at the low voltages is ascribed to the charge injection through the interfacial states. An increase of the voltage causes a gradual filling of states that represents rise of the activation energy. After the filling of all interfacial states the charge is injected only over the interfacial barrier, which stands for second charge relaxation. This assumption is supported by diminishing of interfacial state relaxation after reaching of TFL region. Since steady-state methods are sensitive only to the major bottleneck of the charge injection/transport.

Publication P5: Layer by layer electrical analysis of organic light-emitting diodes

The above-mentioned analysis illustrated the difficulty of the discussion on the origin of the charge relaxation process. To distinguish the origin of the relaxation processes in ITO/ α -NPB/Alq3/Al structure, the ITO/ α -NPB/Al and ITO/Alq3/Al devices have been investigated also. The schematic diagram of the investigated devices is shown in Fig. 1. These results have been presented on the *12th International Conference on Organic Electronics*.

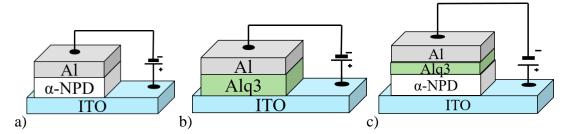


Fig. 1. Sketches of: a) ITO/α-NPB/Al, b) ITO/Alq3/Al, c) ITO/α-NPB/Alq3/Al devices

ITO/ α -NPB/Al device in the voltage range from 2 V to 7 V shows a knee in the log-log scale, with is not appropriate to applied the Child's law evaluation. The difference of this law has been explained by the injection limited $(lnJ \propto \sqrt{V})$ behavior at low voltage region.

ITO/Alq3/Al device opposite to the previously one exhibited pure power dependence of current density on applied voltage bias. This fact pointed out the Child's law, $J \propto V^m$, representing bulk-limited current properties.

The OLED device, ITO/ α -NPB/Alq3/Al, is the most complex structure from all three investigated devices. At the low temperatures the device has a behave as α -NPD device, with injection limited properties, while at the high temperatures dominates Alq3 properties, bulk limited properties.

The comparison of the calculated activation energies from the steady state current-voltage impedance spectroscopy measurements obviously shows almost identical activation energies. According to the energy diagram (Fig. 2) it can be stated that evaluated activation energy of OLED device is dependent on the interface between two organic materials, it is not related to the material of the single organic layer.

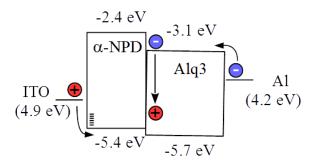


Fig. 2. Energy band diagram of ITO/ α -NPB/Alq3/Al

3 Conclusion

The rapidly expanding field of electronics needs new materials, because the Si and other inorganic semiconductor devise reached the maximum of development. One of the solution way is using the organic semiconductor materials. Application of these new materials gives new electronic devices which have unique properties such as flexible, transparent and etc. Although these devices are in the market, but the charge carrier injection and charge carrier transport is still representing the main challenge of the present applied research. There are many experimentally methods exist to study these physical properties of this layer but not all gives complex information about these phenomena. The goal of the dissertation thesis is finding the correct methods to evaluate the charge carrier injection and transport properties.

The first part of the thesis introduces the fundamentals on organic semiconductors, the charge carrier injection, transport, and defects. As object of this study is finding the appropriate measurement methods and evaluation processes of the study of charge carrier injection and transport in organic semiconductor materials.

The second part of this thesis gives information on three different measurement techniques suggested for characterization charge carrier behavior in organic semiconductors. The steady-state current-voltage measurements reveals the only major contribution of the current limitation. The impedance spectroscopy provides characterization of multiple relaxation processes using frequency-domain measurement. The last but not least is the transient capacitance measurement based on the large-signal method recording the signal in time domain. All three techniques have been used in this thesis for characterization of charge carrier behavior and finding the correlation between these techniques.

Main achievements of the thesis are reported in the third part of the thesis. The brief description of the original results is accompanied with research papers enclosed at the end of this thesis. The attached research papers consist of four journal papers published in international peer-review journals recognized in Master Journal List (the Current Content database) of the Thomson Reuters corporation and one conference contribution presented on the 12th *International Conference on Organic Electronics*.

Organic semiconductors are new materials used for electronic devices. Although they possess some properties of inorganic semiconductors, the charge injection and transport phenomena are often based on

different processes. As a result, the evaluation of charge behaviour in organic semiconductors is still a great challenge. Only the deep understanding to experimental techniques and required approximation allows correct analysis of device properties. Each experimental technique has certain limitations and the data analysis is valid only in a region of applied approximations. This thesis brings a new and original contribution to the evaluation of the organic electronics devices using combination of steady-state and transient methods. The mutual comparison of characterization techniques and detail analysis of obtained results pointed out their strengths as well as limitations. The last but not least contribution of this work is related to the finding that the charge relaxation at the interfaces plays a key role in organic electronics devices and it gives a hint to the technology for further device improvements.

Summary of dissertation thesis

The present work of its contains meets the set objectives. Main achievements of the dissertation thesis can be summarized as follows:

- Acquire new knowledge of organic semiconductors, charge carrier injection, transport and defect states in these materials.
- Acquire skills to prepare organic devices, such as organic diodes, organic filed-effect transistors and organic light-emitting diodes.
- Adjustment of three measurement methods: steady-state current-voltage, impedance spectroscopy and transient capacitance measurement to organic semiconductors.
- Characterizations organic semiconductors by these methods and finding the correlation between these methods.
- Application acquired knowledges measurement for different types of organic semiconductors.
- Identify different charge carrier transport properties in organic materials.
- Analysis of multilayer organic devices: double layer organic light-emitting diodes.

4 Resumé

Neustále sa rozvíjajúci odbor elektroniky si vyžaduje vývoj nových materiálov, nakoľko prvky založené na Si ako aj iných anorganických materiáloch už dosiahli vysoko pokročilú úroveň vývoja, že ďalšie zlepšovanie prvkov je veľmi obtiažne. Jedným z možných riešení sa javí využitie organických polovodičov pre aktívne vrstvy elektronických prvkov. Využitie týchto materiálov dáva elektronickým prvkom nové a unikátne vlastnosti ako je ohybnosť a priehľadnosť a zároveň je možné dosiahnuť výrazne nižšie výrobné náklady. Aj keď niektoré aplikácie sú už komerčne dostupné, tak injekcia a transport náboja stále ostávajú neprekonanou výzvou aplikovaného výskumu. Napriek tomu, že existujú viaceré experimentálne metódy pre charakterizáciu elektrických vlastností prvkov, žiadna metóda neposkytuje komplexný pohľad na fyzikálne javy injekcie a transportu náboja. Cieľom tejto dizertačnej práce je nájdenie korektnej metódy vyhodnocovania injekcie a transport nosičov náboja.

Prvá časť dizertačnej práce sa venuje úvodu do základov organických polovodičov, ako aj problému injekcie, transportu, a záchytu náboja. Druhá časť sa venuje rôznym charakterizačným technikám používaných na vyhodnocovanie elektrických vlastností prvkov. Meranie voltampérových charakteristík v rovnovážnom stave sú schopné odhaliť len najvýznamnejšie javy obmedzujúce tok prúdu naprieč prvkom. Impedančná spektroskopia využíva merania pri rôznych frekvenciách pre poskytnutie informácií o viacerých relaxačných procesoch. Merania tranzientej kapacity sú založené na pozorovaní kapacitnej odozvy v časovej oblasti po aplikovaní veľkého signálu.

Dosiahnuté hlavné výsledky sú zhrnuté v tretej časti práce. Nasleduje stručný opis výsledkov ako aj osobný prínos v publikovaných prácach nachádzajúcich sa v prílohe predloženej dizertačnej práce. Priložené práce boli publikované v štyroch článkoch recenzovaných časopisov evidovaných v Master Journal List (databáza Current Content) spoločnosti Thomson Reuters corporation a jedna práca bola prezentovaná na medzinárodnej konferencii 12th *International Conference on Organic Electronics*.

Organické polovodiče patria medzi nové a perspektívne materiály pre progresívne elektronické prvky a systémy. Aj keď organické polovodiče majú niektoré vlastnosti anorganických polovodičov, mikroskopické príčiny a fyzikálna podstata procesov injekcie a transport náboja sa častokrát odlišuje. Aj preto je vyhodnocovanie správania sa náboja v organických polovodičov ešte stále veľkou výzvou súčasného výskumu. Jedine dôsledné pochopenie charakterizačných technik a teoretických aproximácii vyžadovaných pre vyhodnotenie meraní tak umožní korektnú analýzu vlastností prvkov. Každá experimentálna technika ako aj analýza meraní má svoje vlastné obmedzenia platnosti a vhodnosti využitia. Táto dizertačná práca má snahu o originálny a nový príspevok ku metodike vyhodnocovania prvkov organickej elektroniky pomocou kombinácie rovnovážnych a tranzientných elektrických charakterizačných techník. Vzájomné porovnanie týchto meraní poskytlo nielen informácie o výhodách jednotlivých analýz, ale aj o limitoch ich využitia. Práca taktiež prináša zistenia, že rozhrania materiálov

hrajú kľúčovú úlohu v prvkoch organickej elektroniky a tak dáva nápovedu výrobným technológiám pre ďalšie vylepšovanie prvkov.

Hlavné prínosy dizertačnej práce:

- Získanie nových poznatkov o organických polovodičoch, o injekcií a transporte náboja ako aj o pasciach v týchto materiáloch
- Získanie zručnosti príprave organicky diód, organických poľom riadených
- tranzistoroch a organických elektroluminiscenčných diód.
- Prispôsobenie meracích metód pre merania organických polovodičov: rovnovážnych
- voltampérových meraní, impedančnej spektroskópie a tranzientnej kapacitnej metóde.
- Charakterizácia organických polovodičov týmito metódami a nájdenie zhody medzi týmito metódami.
- Určovanie rôznych typov transport náboja.
- Analýza viacvrstvovej organickej elektroluminiscenčnej diódy.

List of personal publications

ADC Scientific publications in foreign current content papers

- ADC01 <u>JUHÁSZ, Peter</u> [30 %] VÁRY, Michal [10 %] <u>STUCHLÍKOVÁ, Ľubica</u> [20 %] <u>HARMATHA, Ladislav</u> [5 %] <u>JAKABOVIČ, Ján</u> [5 %] <u>WEIS, Martin</u> [30 %]. Characterization of charge traps in pentacene diodes by electrical methods. In *Organic Electronics*. Vol. 17, (2015), s. 240-246. ISSN 1566-1199. In database: CC: 000348495400031.
- ADC02 <u>JUHÁSZ, Peter</u> [20 %] NEVŘELA, Juraj [5 %] MIČJAN, Michal [5 %] NOVOTA, Miroslav [5 %] <u>UHRÍK, Ján</u> [15 %] <u>STUCHLÍKOVÁ, Ľubica</u> [15 %] JAKABOVIČ, Ján [15 %] <u>HARMATHA, Ladislav</u> [5 %] <u>WEIS, Martin</u> [15 %]. Charge injection and transport properties of an organic light-emitting diode. In *Beilstein Journal of Nanotechnology*. Vol. 7, (2016), s. 47-52. ISSN 21904286. In database: CC: 000368133100001.
- ADC03 NEVŘELA, Juraj [12 %] MIČJAN, Michal [12 %] NOVOTA, Miroslav [12 %] KOVÁČOVÁ, Soňa [11 %] PAVÚK, Milan [11 %] JUHÁSZ, Peter [11 %] KOVÁČ, Jaroslav Jr. [11 %] JAKABOVIČ, Ján [10 %] WEIS, Martin [10 %]. Secondary doping in poly(3,4-ethylenedioxythiophene):Poly(4-styrenesulfonate) thin films. In Journal of polymer science. Part B. Polymer physics. Vol. 53, No. 16 (2015), s. 1139–1146. ISSN 0887-6266. In database: CC: 000357287100006.
- ADC04 <u>STUCHLÍKOVÁ, Ľubica</u> [15 %] <u>HARMATHA, Ladislav</u> [15 %] <u>PETRUS, Miroslav</u> [10 %] - <u>RYBÁR, Jakub</u> [15 %] - <u>ŠEBOK, Ján</u> [5 %] - SCIANA, Beata [5 %] -RADZIEWICZ, Damian [5 %] - PUCICKI, Damian [5 %] - TLACZALA, Marek [5 %] -<u>KÓSA, Arpád</u> [5 %] - <u>BENKO, Peter</u> [5 %] - <u>KOVÁČ, Jaroslav</u> [5 %] - <u>JUHÁSZ, Peter</u> [5 %]. Electrical Characterization of the AIIIBV-N Heterostructures by Capacitance Methods. In *Applied Surface Science*. Vol. 269 (2013), s.175-179. ISSN 0169-4332.
- ADC05 <u>STUCHLÍKOVÁ, Ľubica</u> [20 %] <u>WEIS, Martin</u> [20 %] <u>JUHÁSZ, Peter</u> [20 %] <u>KÓSA, Arpád</u> [20 %] <u>HARMATHA, Ladislav</u> [10 %] <u>JAKABOVIČ, Ján</u> [10 %]. Defect Analysis of Pentacene Diode. In *Acta Physica Polonica A : International Conference on Defects Recognition, Imaging and Physics in Semiconductors, Warsaw, Poland, 15-19 Sept. 2013.* Vol. 125, iss. 4 (2014), s. 1038-1041. ISSN 0587-4246.
- ADC06 <u>UHRÍK, Ján</u> [16 %] <u>JAKABOVIČ, Ján</u> [7 %] <u>ŠATKA, Alexander</u> [7 %] VINCZE, Andrej [7 %] <u>FLICKYNGEROVÁ, Soňa</u> [7 %] <u>SLÁDEK, Ľubomír</u> [7 %] <u>KUZMA, Anton</u> [7 %] <u>JUHÁSZ, Peter</u> [7 %] <u>HORÍNEK, František</u> [7 %] <u>RENDEK, Karol</u> [7 %] <u>TELEK, Peter</u> [7 %] <u>DONOVAL, Martin</u> [7 %] <u>WEIS, Martin</u> [7 %]. Effects of substrate condition on calcium corrosion and its role in the calcium test for water vapour transmission rate. In *Corrosion Science*. Vol. 88 (2014), s. 400-404. ISSN 0010-938X.
- ADC07 <u>DONOVAL, Martin</u> [12 %] MIČJAN, Michal [8 %] NOVOTA, Miroslav [8 %] NEVŘELA, Juraj [8 %] KOVÁČOVÁ, Soňa [8 %] PAVÚK, Milan [8 %] JUHÁSZ,
 <u>Peter</u> [8 %] JAGELKA, Martin [8%] KOVÁČ, Jaroslav Jr. [8 %] JAKABOVIČ, Ján [8 %] CIGAN, Marek [8%] WEIS, Martin [8 %]. Relation between secondary doping and phase separation in PEDOT:PSS films. In *Applied Surface Science*. In press. doi:10.1016/j.apsusc.2016.05.076

AFC Published contributions from international scientific conferences

- AFC01 <u>STUCHLÍKOVÁ, Ľubica</u> [40 %] <u>BENKOVSKÁ, Jana</u> [20 %] <u>JAKUŠ, Juraj</u> [10 %] <u>HARMATHA, Ladislav</u> [10 %] <u>NEMEC, Michal</u> [5 %] <u>BENKO, Peter</u> [5 %] <u>KÓSA, Arpád</u> [5 %] <u>JUHÁSZ, Peter</u> [5 %]. DLTFS Study of Deep Traps in Schottky Diodes Based on AlGaN/GaN Heterostructure. In *EDS'13. Electronic Devices and Systems IMAPS CS International Conference 2013 : Proceedings; Brno, Czech Republic, 26-27 June 2013. Brno : Vysoké učení technické v Brně, 2013, s.98-103. ISBN 978-80-214-4754-7.*
- AFC02 <u>STUCHLÍKOVÁ, Ľubica</u> [30 %] <u>BENKOVSKÁ, Jana</u> [15 %] <u>NEMEC, Michal</u> [10 %] <u>KÓSA, Arpád</u> [10 %] <u>JUHÁSZ, Peter</u> [10 %] <u>JAKUŠ, Juraj</u> [10 %] <u>BENKO, Peter</u> [15 %]. E-Learning in Engineering Education. In Nové technologie ve výuce [elektronický zdroj] : 6. ročník mezinárodní konference. Postkonferenční sborník abstraktů a elektronických verzí příspěvků. Brno, 15. 11. 2012. Brno : Masarykova univerzita, 2013, s.CD-ROM, [5] s. ISBN 978-80-210-6402-7.

AFD Published contributions from domestic scientific conferences

- AFD01 JUHÁSZ, Peter [50 %] PETRUS, Miroslav [25 %] STUCHLÍKOVÁ, Ľubica [25 %]. Identifikácia porúch v GaAsN štruktúrach spektroskopiou hlbokých hladín. In ŠVOČ 2012 [elektronický zdroj] : Zborník vybraných prác, Bratislava, 25. apríl 2012. Bratislava : FEI STU, 2012, s.CD-ROM, s. 321-325. ISBN 978-80-227-3697-8.
- AFD02 <u>JUHÁSZ, Peter</u> [40 %] <u>WEIS, Martin</u> [25 %] <u>STUCHLÍKOVÁ, Ľubica</u> [20 %] -<u>PETRUS, Miroslav</u> [5 %] - <u>JAKABOVIČ, Ján</u> [5 %] - <u>HARMATHA, Ladislav</u> [5 %]. Investigation of Emission Processes in Pentacene. In *ADEPT 2013 : 1st International Conference on Advances in Electronic and Photonic Technologies. Nový Smokovec, High Tatras, Slovakia, June 2-5, 2013.* 1. vyd. Žilina : University of Žilina, 2013, s.127-130. ISBN 978-80-554-0689-3.
- AFD03 JUHÁSZ, Peter [40 %] STUCHLÍKOVÁ, Ľubica [30 %] WEIS, Martin [30 %].
 Štúdium emisných procesov v pentacéne metódou DLTS. In ŠVOČ 2013 [elektronický zdroj] : Zborník vybraných prác, Bratislava, 23. apríl 2013. 1. vyd. Bratislava : FEI STU, 2013, s.CD ROM, s. 309-312. ISBN 978-80-227-3909-2.
- AFD04 JUHÁSZ, Peter [30 %] STUCHLÍKOVÁ, Ľubica [20 %] MIČJAN, Michal [20 %] HARMATHA, Ladislav [5 %] JAKABOVIČ, Ján [5 %] WEIS, Martin [20 %].
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- AFD05 <u>JUHÁSZ, Peter</u> [35 %] VÁRY, Michal [10 %] <u>STUCHLÍKOVÁ, Ľubica</u> [20 %] <u>JAKABOVIČ, Ján</u> [10 %] <u>HARMATHA, Ladislav</u> [5 %] <u>WEIS, Martin</u> [20 %]. Defects characterization in a pentacene diode structure by different electrical methods. In *ADEPT* 2014 : 2nd International Conference on Advances in Electronic and Photonic Technologies; Tatranská Lomnica, Slovakia; June 1-4, 2014. 1.vyd. Žilina : University of Žilina, 2014, s. 116-119. ISBN 978-80-554-0881-1.

- AFD06 <u>JUHÁSZ, Peter</u> [55 %] GERLING, L. G. [3 %] <u>WEIS, Martin</u> [40 %] PUIGDOLLERS, J. [2 %]. Heterojuntion solar cells based on MoO3/c-Si. In ADEPT 2015 : 3rd international conference on advances in electronic and photonic technologies. Štrbské Pleso, High Tatras, Slovakia. June 1-4, 2015. 1. vyd. Žilina : University of Žilina, 2015, S. 160-163. ISBN 978-80-554-1033-3.
- AFD07 <u>KÓSA, Arpád</u> [25 %] <u>JUHÁSZ, Peter</u> [25 %] <u>BENKOVSKÁ, Jana</u> [25 %] <u>STUCHLÍKOVÁ, Ľubica</u> [25 %]. Deep Level Investigation on AlGaN/GaN Structures by DLTS Method. In ŠVOČ 2013 [elektronický zdroj] : Zborník vybraných prác, Bratislava, 23. apríl 2013. 1. vyd. Bratislava : FEI STU, 2013, s.CD ROM, s. 319-324. ISBN 978-80-227-3909-2.
- AFD08 MIČJAN, Michal [25 %] NEVŘELA, Juraj [15 %] NOVOTA, Miroslav [15 %] -<u>FLICKYNGEROVÁ, Soňa</u> [5 %] - <u>JUHÁSZ, Peter</u> [10 %] - <u>UHRÍK, Ján</u> [5 %] -<u>JAKABOVIČ, Ján</u> [10 %] - <u>WEIS, Martin</u> [15 %]. Ferroelectric polymer films for flexible memory devices. In ASDAM 2014 : The 10th International Conference on Advanced Semiconductor Devices and Microsystems. Smolenice Castle, Slovak Republic, October 20-22, 2014. 1.vyd. Danvers : IEEE, 2014, s. 93-96. ISBN 978-1-4799-5474-2.
- AFD09 NEVŘELA, Juraj [25 %] MIČJAN, Michal [15 %] NOVOTA, Miroslav [10 %] -<u>FLICKYNGEROVÁ, Soňa</u> [10 %] - <u>KOVÁČ, Jaroslav Jr.</u> [10 %] - PAVÚK, Milan [5 %] -<u>JUHÁSZ, Peter</u> [5 %] - <u>JAKABOVIČ, Ján</u> [10 %] - <u>WEIS, Martin</u> [10 %]. Technology of conductive polymer PEDOT:PSS films. In ASDAM 2014 : The 10th International Conference on Advanced Semiconductor Devices and Microsystems. Smolenice Castle, Slovak Republic, October 20-22, 2014. 1.vyd. Danvers : IEEE, 2014, s. 29-32. ISBN 978-1-4799-5474-2.
- AFD10 NOVOTA, Miroslav [20 %] MIČJAN, Michal [10 %] NEVŘELA, Juraj [10 %] -<u>FLICKYNGEROVÁ, Soňa</u> [10 %] - <u>JUHÁSZ, Peter</u> [10 %] - MIŠICÁK, Róbert [10 %] -PUTALA, Martin [10 %] - <u>JAKABOVIČ, Ján</u> [10 %] - <u>WEIS, Martin</u> [10 %]. New phenanthrene-based organic semiconductor material for electronic devices. In ASDAM 2014 : The 10th International Conference on Advanced Semiconductor Devices and Microsystems. Smolenice Castle, Slovak Republic, October 20-22, 2014. 1.vyd. Danvers : IEEE, 2014, s. 97-100. ISBN 978-1-4799-5474-2.
- AFD11 <u>PETRUS, Miroslav</u> [30 %] <u>STUCHLÍKOVÁ, Ľubica</u> [10 %] <u>RYBÁR, Jakub</u> [5 %] <u>JUHÁSZ, Peter</u> [10 %] <u>HARMATHA, Ladislav</u> [5 %] <u>BENKO, Peter</u> [5 %] <u>KOVÁČ, Jaroslav</u> [5 %] <u>ŽIŠKA, Milan</u> [5 %] <u>ŠEBOK, Ján</u> [5 %] ŚCIANA, Beata [5 %] PUCICKI, Damian [5 %] TLACZALA, Marek [5 %] RADZIEWICZ, Damian [5 %]. Deep Level Investigation on GaAsN Structures by DLTS Method. In *APCOM 2012. Applied Physics of Condensed Matter : Proceedings of the 18th International Conference. Štrbské Pleso, Slovak Republic, June 20-22, 2012. 1. vyd. Bratislava : STU v Bratislave, 2012, s.353-356. ISBN 978-80-227-3720-3.*
- AFD12 <u>STUCHLÍKOVÁ, Ľubica</u> [30 %] <u>KÓSA, Arpád</u> [20 %] <u>HARMATHA, Ladislav</u> [10 %] <u>JUHÁSZ, Peter</u> [10 %] <u>KOVÁČ, Jaroslav</u> [10 %] SCIANA, Beata [5 %] DAWIDOWSKI, Wojciech [5 %] PUCICKI, Damian [5 %] TLACZALA, Marek [5 %]. The influence of growth parameters on Emission and capture processes in InGaAsN structures. In *ADEPT 2015 : 3rd international conference on advances in electronic and photonic technologies*. *Štrbské Pleso, High Tatras, Slovakia. June 1-4, 2015*. 1. vyd. Žilina : University of Žilina, 2015, S. 316-319. ISBN 978-80-554-1033-3.

- AFD13 <u>WEIS, Martin</u> [12 %] NEVŘELA, Juraj [11 %] MIČJAN, Michal [11 %] NOVOTA, Miroslav [11 %] <u>FLICKYNGEROVÁ, Soňa</u> [11 %] <u>KOVÁČ, Jaroslav Jr.</u> [11 %] PAVÚK, Milan [11 %] <u>JUHÁSZ, Peter</u> [11 %] <u>JAKABOVIČ, Ján</u> [11 %]. The role of doping in high-conductive PEDOT:PSS films. In *ADEPT 2014 : 2nd International Conference on Advances in Electronic and Photonic Technologies; Tatranská Lomnica, Slovakia; June 1-4, 2014*. 1.vyd. Žilina : University of Žilina, 2014, s. 1-4. ISBN 978-80-554-0881-1.
- AFD14 WEIS, Martin [40 %] KOVÁČOVÁ, Soňa [20 %] JUHÁSZ, Peter [20 %] OTSUKA, Takako [5 %] TAGUCHI, Dai [5 %] MANAKA, Takaaki [5 %] IWAMOTO, Mitsumasa [5 %]. Probing of the charge behaviour in organic light-emitting diode with PEDOT:PSS anode. In ADEPT 2015 : 3rd international conference on advances in electronic and photonic technologies. Štrbské Pleso, High Tatras, Slovakia. June 1-4, 2015. 1. vyd. Žilina : University of Žilina, 2015, S. 80-83. ISBN 978-80-554-1033-3.

AFG Abstracts of contribution from international conferences

AFG01 <u>STUCHLÍKOVÁ, Ľubica</u> [40 %] - <u>HARMATHA, Ladislav</u> [10 %] - <u>PETRUS, Miroslav</u>
[5 %] - <u>RYBÁR, Jakub</u> [5 %] - <u>ŠEBOK, Ján</u> [5 %] - KÓSA, Karol [5 %] - <u>BENKO, Peter</u> [5 %] - <u>KOVÁČ, Jaroslav</u> [5 %] - <u>JUHÁSZ, Peter</u>. Electrical Characterization of AIIIBv-N
Heterostructures by Capacitance Methods. In *SURFINT - SREN III : Progress in Applied Surface, Interface and Thin Film Science 2012. Florence, Italy, May 14-18, 2012.* Bratislava : Comenius University, 2012, s.180-181. ISBN 978-80-223-3212-5.

BEF Scientific works in domestic proceedings

- BEF01 JUHÁSZ, Peter [35 %] STUCHLÍKOVÁ, Ľubica [15 %] JAGELKA, Martin [5 %] DONOVAL, Martin [5 %] DAŘÍČEK, Martin [5 %] WEIS, Martin [35 %]. Study of charge traps in organic diodes by steady-state and transient techniques. In WOCSDICE 2015 : 39th workshop on compound semiconductor devices and integrated circuits. Smolenice, Slovakia. 8-10 June, 2015. Bratislava : UEF FEI STU, 2015, p. 35-36.
- BEF02 JUHÁSZ, Peter [12 %] UHRÍK, Ján [8 %] MIČJAN, Michal [8 %] NEVŘELA, Juraj [8 %] NOVOTA, Miroslav [8 %] WEIS, Martin [8 %].- STUCHLÍKOVÁ, Ľubica [8 %] Vary, Michal [8 %]. Layer by layer electrical analysis of organic light emitting diodes OLED. In ICOE 2016 [elektronický zdroj] : Book of Abstracts. 12th International Conference on Organic Electronics. Bratislava, Slovakia, June 13-15, 2016. Bratislava, p. 36-37.

BFA Abstracts of scientific works from international conferences

BFA01 JUHÁSZ, Peter [20%] - VÁRY, Michal [16%] - STUCHLÍKOVÁ, Ľubica [16%] - JAKABOVIČ, Ján [16%] - HARMATHA, Ladislav [16%] - WEIS, Martin [16%]. Electrical characterization of pentacene diode by complementary techniques. In ICOE 2014 [elektronický zdroj] : Book of Abstracts. 10th International Conference on Organic Electronics. Modena, Italy, June 11-13, 2014. Modena : Universita di Modena, 2014, online, [2] p.

- BFA02 JUHÁSZ, Peter [20 %] NEVŘELA, Juraj [10 %] MIČJAN, Michal [10 %] NOVOTA, Miroslav [10 %] - <u>UHRÍK, Ján</u> [10 %] - <u>STUCHLÍKOVÁ, Ľubica</u> [10 %] - <u>HARMATHA,</u> <u>Ladislav</u> [5 %] - <u>JAKABOVIČ, Ján</u> [5 %] - <u>WEIS, Martin</u> [20 %]. Charge transport properties of organic light emitting diode. In ECOF-14 [elektronický zdroj] : 14th European conference on organized films. Genova, Italy. June 29 - July 2, 2015. Genova : University of Genova, 2015, USB, [O69].
- BFA03 JUHÁSZ, Peter [40 %] VÁRY, Michal [10 %] STUCHLÍKOVÁ, Ľubica [20 %] WEIS, Martin [30 %]. Charge relaxation times in pentacene diode estimated by steardy-state and transient methods. In ICDS 2015 [elektronický zdroj] : 28th International conference on defects in semiconductors. Espoo, Finland. July 27 – 31, 2015. Espoo : Aalto University, 2015, USB, [2] s.
- BFA04 WEIS, Martin [16 %] FLICKYNGEROVÁ, Soňa [14 %] NEVŘELA, Juraj [14 %] MIČJAN, Michal [14 %] NOVOTA, Miroslav [14 %] JUHÁSZ, Peter [14 %] JAKABOVIČ, Ján [14 %]. High-conductive PEDOT:PSS films: The true role of doping. In ICOE 2014 [elektronický zdroj] : Book of Abstracts. 10th International Conference on Organic Electronics. Modena, Italy, June 11-13, 2014. Modena : Universita di Modena, 2014, online, [2] p.

GII Other publications

GII01 <u>STUCHLÍKOVÁ, Ľubica</u> [20 %] - <u>WEIS, Martin</u> [20 %] - <u>JUHÁSZ, Peter</u> [20 %] - <u>KÓSA, Arpád</u> [20 %] - <u>HARMATHA, Ladislav</u> [10 %] - <u>JAKABOVIČ, Ján</u> [10 %]. Deep Level Transient Spectroscopy Study of Pentacene Diode. In 15 th International Conference on Defects Recognition, Imaging and Physics in Semiconductors : Warsaw, Poland, September 15-19, 2013, 2013, s.80-81.

Statistic of citation

Citation in foreign papers, registered in Web of Science and SCOPUS databases 3

ADC03 NEVŘELA, Juraj [12 %] - MIČJAN, Michal [12 %] - NOVOTA, Miroslav [12 %] - KOVÁČOVÁ, Soňa [11 %] - PAVÚK, Milan [11 %] - JUHÁSZ, Peter [11 %] - KOVÁČ, Jaroslav Jr. [11 %] - JAKABOVIČ, Ján [10 %] - WEIS, Martin [10 %]. Secondary doping in poly(3,4-ethylenedioxythiophene):Poly(4-styrenesulfonate) thin films. In Journal of polymer science. Part B. Polymer physics. Vol. 53, No. 16 (2015), s. 1139–1146. ISSN 0887-6266. In database: CC: 000357287100006.

1. DIAH AWM, QUIRINO JP, BELCHER W, HOLDSWORTH CI, "An Assessment of the Effect of Synthetic and Doping Conditions on the Processability and Conductivity of Poly(3,4-ethylenedioxythiophene)/Poly(styrene sulfonic acid)", *Macromolecular Chemistry and Physics*, in press (2016).

2. CHEN L, QIU S, LIU P, XIONG F, LU J, LIU Y, LI G, LIU Y, REN F, XIAO Y, GAO L, ZHAO Q, DING B, LI Y, GUO Y, CHEN X, "The description of charge transfer in fast negative ions scattering on water covered Si(100) surfaces", *Applied Surface Science*, in press (2016).