



VYSOKÉ UČENÍ TECHNICKÉ V BRNĚ

BRNO UNIVERSITY OF TECHNOLOGY



FAKULTA ELEKTROTECHNIKY A KOMUNIKAČNÍCH
TECHNOLOGIÍ

ÚSTAV MIKROELEKTRONIKY

FACULTY OF ELECTRICAL ENGINEERING AND COMMUNICATION
DEPARTMENT OF MICROELECTRONICS

STUDY OF ORGANIC BASED STRUCTURES FOR SOLAR CELLS

STUDIUM STRUKTUR PRO ORGANICKÉ FOTOVOLTAICKÉ SOLÁRNÍ ČLÁNKY

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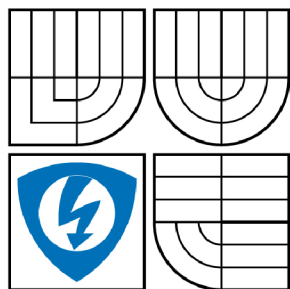
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VEDOUCÍ PRÁCE

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VYSOKÉ UČENÍ
TECHNICKÉ V BRNĚ

Fakulta elektrotechniky
a komunikačních technologií

Ústav mikroelektroniky

Bakalářská práce

bakalářský studijní obor

Mikroelektronika a technologie

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NÁZEV TÉMATU:

Studium struktur pro organické fotovoltaické solární články

POKYNY PRO VYPRACOVÁNÍ:

Seznamte se s technologií organických solárních článků. Seznamte se s přípravou tenkovrstvé planární struktury z dodaného organického polovodiče. Charakterizujte materiál z hlediska morfologie, krystalinity, vodivosti a fotovodivosti v koplanárním, případně sendvičovém uspořádání.

DOPORUČENÁ LITERATURA:

- [1] WEITER, M. Latest achievements in organic semiconductors for future electronic devices. In Proceedings of IMAPS International Conference. Brno: IMAPS, 2006. s. XV (6 s.) ISBN: 80-214-3246-2.
[2] SALYK, O.; WEITER, M. Polysilane Luminescent Materials. In Proc. of 11th Conference Electronic Device and Systems 2004. 1. Brno: VUT v Brně, 2004. s. 278-284. ISBN: 80-214-2701-9.

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Vedoucí práce: doc. Ing. Jaroslav Boušek, CSc.

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Vedoucí/školicel VŠKP: doc. Ing. Jaroslav Boušek, CSc.

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Abstrakt

Práce se zabývá problematikou využití organických materiálů v solárních článcích. V úvodu práce jsou rozebrány a vysvětleny jednotlivé fyzikální a chemické jevy, které ovlivňují vlastnosti organických materiálů. Práce pokračuje rozбором působení rozličných struktur organických solárních článků na jejich vlastnosti. Dále je popsána technologie výroby vzorku a jsou v ní uvedeny výsledky měření fotovodivosti.

Abstract

The work deals with the issue of organic materials used in solar cells. The beginning of the analysis and explains physical and chemical effects that influence parameters of organic materials. In addition, the work examines the different solar cell structures and their influence on the performance of the solar cell. At the end of the work, there is an explanation of the technology used for preparing samples and the results of measuring the photoconductivity.

Klíčové slová

Organické polovodičové materiály, solární články, koplanární struktura, sendvičová struktura, vodivost, fotovodivost

Key words

Organic semiconductor materials, solar cells, coplanar structure, laminated structure, conductivity, photoconductivity

Bibliografická citace díla:

- [1] KOSTOVČÍK, T. Studium struktur pro organické fotovoltaické solární články. Brno: Vysoké učení technické v Brně, Fakulta elektrotechniky a komunikačních technologií, 2008. 30 s. Vedoucí bakalářské práce doc. Ing. Jaroslav Boušek, CSc.

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V Brně dne 29. 5. 2008

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Introduction

At the present time, electricity is very important part of our everyday life. We cannot even imagine our life without refrigerator, TV, microwave, computers etc. Consumption of electric energy is very closely connected with life quality. The world trend is that consumption of electricity will be in the future still bigger and bigger, so production of electric energy is very important task for engineers and a lot of researches are focused on improving it. In the past century most of the electric energy has been produced in power plants based on combustion of fossil fuels. Latter when nuclear fusion was invented, the possibility of electricity production in nuclear plants appeared and become very popular in many countries. Nowadays humankind attempts to find also alternative sources of electrical energy and research is focused on wind energy, biomass combustion and solar energy. Each of this production technology has some pros and cons.

Solar energy has shown that it can be used in different situation and applications. It can produce electricity for isolated settlements, far away from electric grids or it can provide electricity for remote transmitters where diesel generators could not be used. It can support electric networks in big cities during summer peak loads, when all air conditioning are on or generate electric energy for satellites that are in outer space for many years. Moreover, the advantage of solar cell is that they do not produce emission. There is no combustion or radioactive fuel disposal. They have low operating cost and there are no moving part so wearing is very low so they are very reliable, because they can operate for over 20 years. They are sensitive for UV degradation. The conversion efficiency goes down rapidly with exception of the most expensive silicon cells.

All of this is possible thanks to photovoltaic technology. Photovoltaic is the technology that generates direct current (DC) electrical power from semiconductors when they are illuminated by photons. As long as light is shining on the solar cell (the name for the individual PV element), it generates electrical power. When the light stops, the electricity stops. Solar cells never need recharging like a battery. Solar cell is intended for solar energy conversion, while photovoltaic cells include also light detectors in technology, cells for calculators etc. [1]

The history of photovoltaic begins when Antoine Henri Becquerel discovered in 1839 photo galvanic effect in liquid electrolytes. The first large area solar cell using Se film was produced by Charles Fritts in 1883. Later NASA research and oil crisis in 1973 influence the photovoltaic technology mostly and bring it to commercial world markets. Nowadays we can divide solar cells technology into 3 generations.

The first generation photovoltaic cell consists of a large-area, single-crystal, single layer p-n junction diode, more frequently p-i-n junction, capable of generating usable electrical energy from light sources with the wavelengths of sunlight. First-generation photovoltaic cells are the dominant technology in the commercial production of solar cells. [1] It is the most efficient conversion, up to 25 %, the longest lifetime, but the highest cost initial investment.

The second generation of photovoltaic materials is based on the use of thin epitaxial deposits of semiconductors on lattice-matched wafers. [1]

The third-generation photovoltaic is proposed to be very different from the previous semiconductor devices, as they do not rely on a traditional p-n junction to separate photo generated charge carriers. For space applications, quantum well devices (quantum dots, quantum ropes, etc.) and devices incorporating carbon nanotubes are being studied - with a potential for up to 45% AM0 production efficiency , where AM0 refers to the spectral distribution of sun radiation intensity just above the Earth's atmosphere. [1]

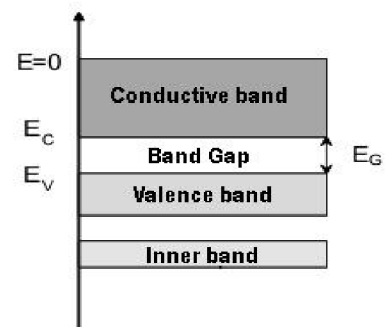
Theoretical part

Some organic materials with double (π) bonded carbon, which behave as semiconductors, can be used for transformation solar energy to electrical. Even if their performance is not as good as performance of inorganic materials, they have one big advantage in simple manufacturing. In organic materials is generation of charge carries much more complicated than in inorganic. Crystal lattice does not consist of simple atoms, but it is created by complicated molecules, which are influenced by different intermolecular forces. There are different chemical bonds between molecules and the distances between molecules are bigger so the band gap is also wider and the mobility of charge carriers is low. Band theory, which explains behavior of inorganic materials, should be helpful but it does not explain all effects, which are in organic materials.

Band Theory

In 1913, Bohr formulates three rules that explain behavior of electrons. There are discrete energy levels in isolated atom and electron can move only on this energy levels. There are attractive and repulsive forces, during interactions of two atoms that depend on a distance of these atoms. In some distance this forces are equal. This distance is called crystal lattice constant. In this status, energy levels are splitted. In bigger amount of atoms, these splitted energy levels create bands as we can use band theory. We recognize four kinds of bands:

1. Inner(filled) bands – electrons in this bands are strongly attracted to the core(they have no influence on electric conductivity)
2. Valence band – electrons in valence band create a chemical bond
3. Conducive band –electrons in this band effect electric conductivity
4. Band gaps – this band separates allowed energy bands. for $T=300\text{K}$; Ge $a=0.565\text{nm} \rightarrow E_g=0.66\text{eV}$; Si $a=0.543 \rightarrow E_g=1.12\text{eV}$)



Picture 1 - Simple band model

Band theory can explain electric conductivity of materials and their division on insulators, semiconductors and conductors. Insulators have full valance band, big band gap (min 3eV) and empty conductive band. Semiconductors have similar structure like isolators but band gap is smaller and small band gap allows transfer of electrons from valence band to conductive band. There are two types in metals. Monocovalent metals have small band gap

and partially fulfilled conductive band. Bivalent metals does not have band gap and conductive and valence band overlay.

Fermi Level

Fermi level is energy level which is occupied with probability 50%. The position of Fermi level can show us two important things about semiconductor.

1. The position of Fermi level can help us to specify kind of conductivity. If the Fermi level is near to conductive band it means that electron are major charge carriers. On the other side, if Fermi level is nearer to valence band it means that the holes are major charge carriers.
2. The position of Fermi level is major factor, which can say us what kind of contact, ohmic or blocking, will become into existence on metal semiconductor interface.

When we apply external voltage on semiconductor, Fermi level divides into 2 separate levels called quazi Fermi levels.

Doping

In organic use of compounds is preferred against elements in doping process. Atoms are too small and can easily move through the lattice and recombine. Another possibility of doping is chemical oxidation and recombination. We can also change concentration of charge carriers by trapping electrons in conductive band. This will move down Fermi level and change conductivity type if semiconductor.

Typical p-type dopants:

- Cl_2NO_2
- Br_2
- H_2O
- O_2
- $TCNQ$
- $C60$

Typical n-type dopants:

- Alkali metals
- Phenothiazine

Photo generation

In organic semiconductor, absorption of photon does not lead to generation of free charge carriers, like in inorganic semiconductors, but to generation of exciton. Exciton need to be dissociated in dissociation centre and separated hole and electron can be injected into external circuit. There are processes from absorption of photo injection into external circuit, which are connected with energy losses.

Absorption of light

- Band gap must be small
- Layer mustn't be thin
- Small reflection of transparent layer is demanded

Exciton diffusion

- Exciton have to have enough diffusion length to reach dissociation centre which can be situated on another side of structure

Charge transport

- Can be affected by recombination
- Interaction with lattice and other atoms can slow down the transport speed

Dissociation centre

There are different ways how to dissociate exciton and create separated holes and electrons. Some are effective, like metal semiconductor interface and D/A interface, and some are less effective, like impurities and oxygen traps.

Oxygen traps

Electron can be trapped in oxygen trap. After certain time its energy decreases and it falls out from oxygen trap into valence band, where it can recombine with free hole or hole of an exciton. This effect is possible but not very efficient and oxygen can have other negative effects on abilities of material.

D/A interface

If material has big electron affinity it can easily accept electron. This material is called electron acceptor (A). On the other side if a material has a low ionization energy it can easily accept hole, and donate electron to A, so they are called donors (D). If exciton gets on interface of acceptor and donor, material with big electron affinity can attract electron without difficulty and material with low ionization energy can accept hole. This leads into separation of free charge carriers from exciton. Energy difference between electron affinity and ionization energy has to be enough big otherwise it will not lead into exciton splitting and it will simply pass through interface and recombine.

Energy Losses in Solar Cells

The processes in organic materials from absorption of solar energy until injection into outer electric circuit are complicated and they are connected with energy losses, which need to be eliminated or minimal.

Material reflection and through pass

The losses created by reflection should be very significant, but there are only limited possibilities how to reduce them. One of the most common is usage of antireflection layers, which are already used in inorganic solar cells.

If the band gap is big, the change that photon energy will not be absorbed is growing, and photon will simply pass through material. For semiconductor polymers is band gap

usually about 2eV. This limits the absorption to about 30%. This effect should be easily reduced by using a reflexive material for a back contact, which reflects the solar energy back through material, where it had change to interact again.

Exciton recombination

Exciton is created by connection of electron and hole pair, which have to travel to contacts, where they will be injected into outer electric field. If the transport time to dissociation centre is bigger than their lifetime, they will be recombined.

Recombination of charge carriers.

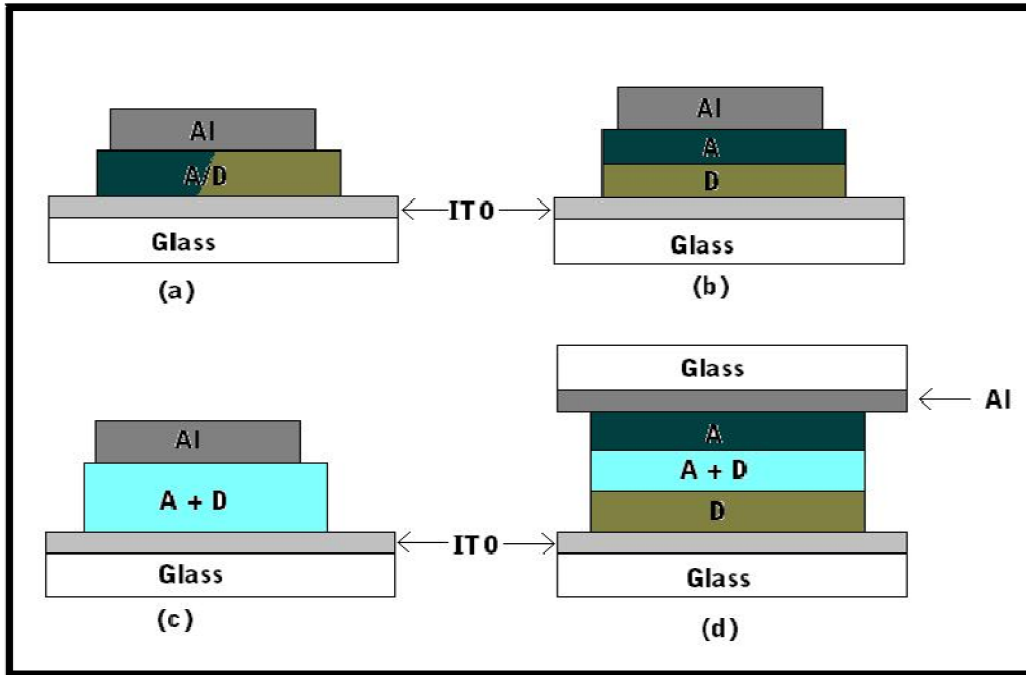
If exciton dissociate in dissociation centre, the free charge carriers (electron and hole) are created, and they have to travel to contacts, where they will be injected into outer electric field. If the excitons transport time to dissociation centre is bigger than their lifetime, they will recombine.

Barriers on contacts

If the charge carriers reach the contact with low work function, it has to pass thin oxide layer, which create a potential barrier. There should be also created an ohmic contact on side, which limits the mean free path of carriers.

Structures of organic solar cells

With the development of organic solar cells the few different structures has been invented. Each of them has its own pros and cons.



Picture 2 - Structures of organic solar cells (a) single layer (b) double layer (c) blend (d) sandwich

Single layer structure

Single layer structure is the simplest form of organic solar cells. It consists of transparent antireflexive substrate (glass), indium tin oxide transparent contact (ITO), absorbing organic film, reflective Al contact. Accidental photons get through glass and ITO some of them create excitons in organic film another part get to the back electrode and they are reflected back through organic film where they have another possibility to create exciton and some of them get out of structure without interaction. The organic film is from 40nm to 200nm thick. Most important dissociation centre in this structure is metal (Al)-semiconductor interface also ITO/semiconductor may become active.

Double layer structures

With introduction of an acceptor layer between the active material and negative electrode, the exciton diffusion length can be improved. In addition, difference of electron affinity in electron acceptor and ionization energy in electron donor material can split exciton and separated charge carriers can get to electrode with smaller change to meet recombination partner. The active region is now extended to the exciton diffusion length of both materials. The possibility of choice of two organic materials gives us change to cover demanded spectrum scale.

Effects that happen on D/A interface are very similar to this at p/n junction in inorganic semiconductors. In both types equalization of Fermi level occurs. If the difference between electron affinity and ionization energy is not sufficient exciton just passes through the

interface without splitting and very probably recombine. Possibility to optimize organic double layer cells is to place the material with the higher band gap next to transparent contact and the lower band gap material behind, but there is necessary to take care about second metal semiconductor contact to avoid creation of blocking contact.

Blend layer structures

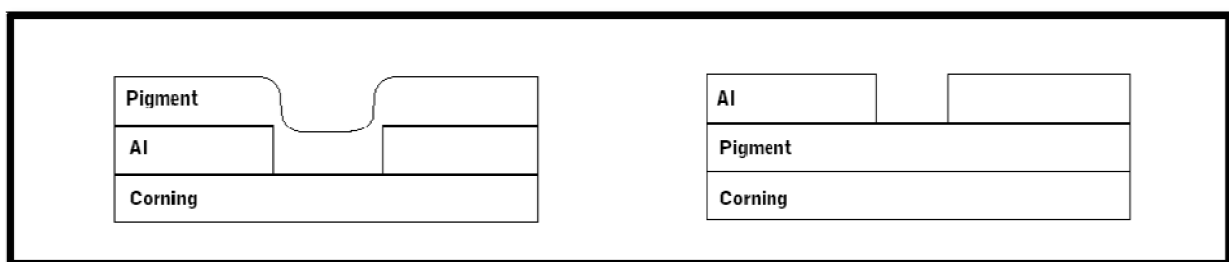
Blend layer structure use organic material, which is mixture of donor and acceptor. Because in structure are both materials they can form a charge separating interface and the film can be thicker. One of advantage over double layer is that, under some conditions, active layer can be thicker than the sum of the exciton diffusion lengths. This means that they can accept 15times more photons then typical single layer structures. Despite the large active region, most blends devices develop lower open circuit voltages than single layer structures – reasons are not well understand yet.

Laminated structures

Another possibility to construct multi layer solar cells are laminated solar cell. The idea is to deposit both acceptor and donor material on separated substrate/electrode and then join them to get D/A interface. This could be very useful when we need to treat each organic material separately. Such D/A interface have much better performance than in double layer or in blend structure. The properties of the D/A interface can be controlled by heating, applying a pressure or other processes during or after fabrication.

Experimental part

One of the possible structures for characterization of material is coplanar structure. There are two possible variants for these structures (picture 3)

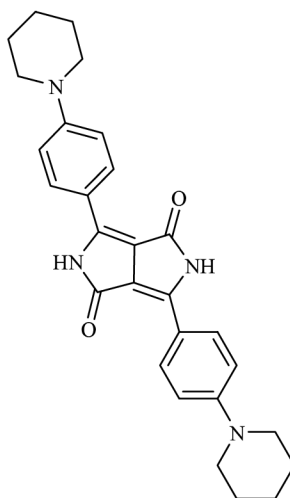


Picture 3 - Coplanar structure for current versus voltage measurement

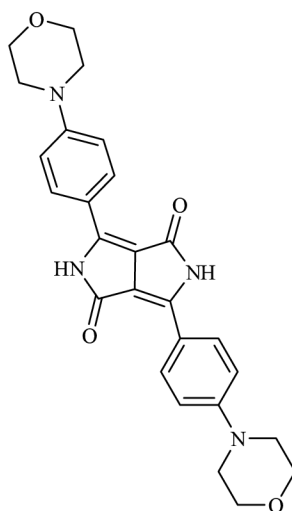
Low alkali Corning glass 7059 is used as bearing material for structure, pigment is organic material, which is examined and aluminum is used for a contacts. The layer of aluminum and organic materials are created by vapour deposition through the mask.

Materials

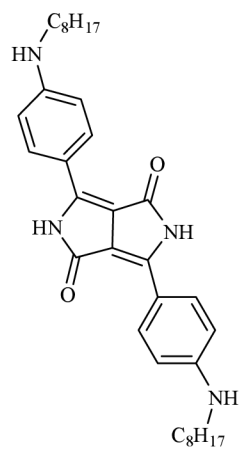
For our measuring, we used PPV – Fullerene as the reference material. This material is already used in commercial applications and in our measuring is used as reference material. We prepared two samples, one prepared by spin coating method and another one prepared by simple dropping under angle of about 45°. From our materials, we decided to use material marked as U14, U15, U16 and U29.



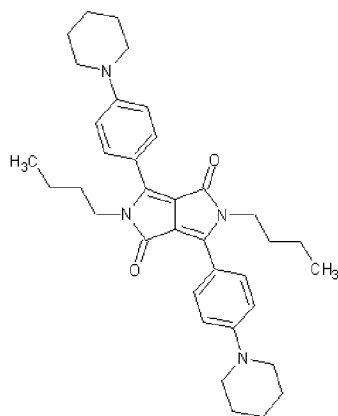
Picture 4 - U14



Picture 5 - U 15



Picture 6 - U16



Picture 7 - U29

Vacuum technology

Vacuum technology allows the particles to move more freely, because amount of other particles are reduces and mean free path is increased. Vacuum technology also allows that processes in the object and on their surface proceed without undesirable effects. For example tungsten filament in standard conditions get burn within 1 minute, because on one cm^2 of his surface, impact 10^{23} molecules of oxygen per 1 second. In the vacuum with pressure 10^{-5} Pa is it only about 10^{13} molecules per second, so the filament does not get burned so rapidly. Physical principle of vapour deposition is liberation of molecules deposited material evoked by heating. Probability that the particle of deposited material reach the point x with distance d is get by formula

$$p \cong \left[1 - e^{-\frac{d}{\lambda}} \right] \quad (1)$$

It is clear that mean free path of molecule λ depends on pressure, shown in the table 1.

Pressure [Pa]	10^1	10^{-1}	10^{-3}	10^{-5}	10^{-7}
λ [cm]	0.5	51	510	5.1×10^4	5.1×10^6

Table 1 - Average free movement versus pressure

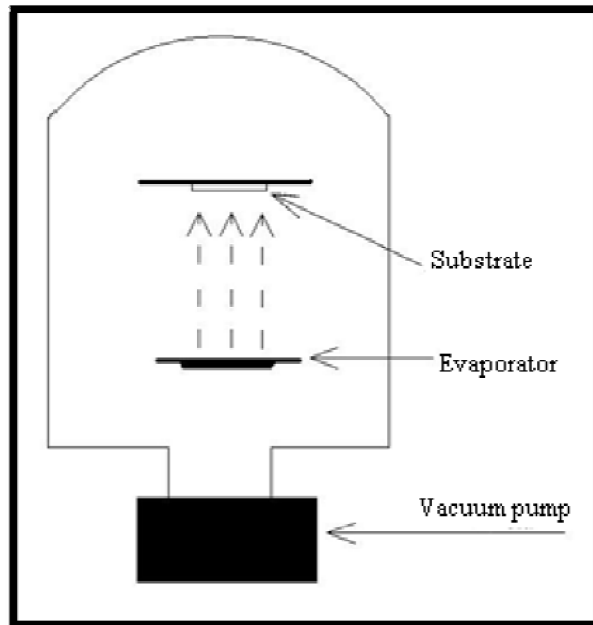
Amount of particles N deposited per time unit from area unit is get by formula:

$$N = 3,5 \cdot 10^{22} \times \frac{p}{(M \times t)^{-\frac{1}{2}}} \quad (2)$$

Where

- p pressure
- M molecular weight
- t temperature

Vacuum deposition system consist of 3 main parts, deposition chamber, vacuum pump (usually system of pumps) and evaporator



Picture 8 - Model of vapour depositor



Picture 9 - Vapour depositors

Vacuum pumps

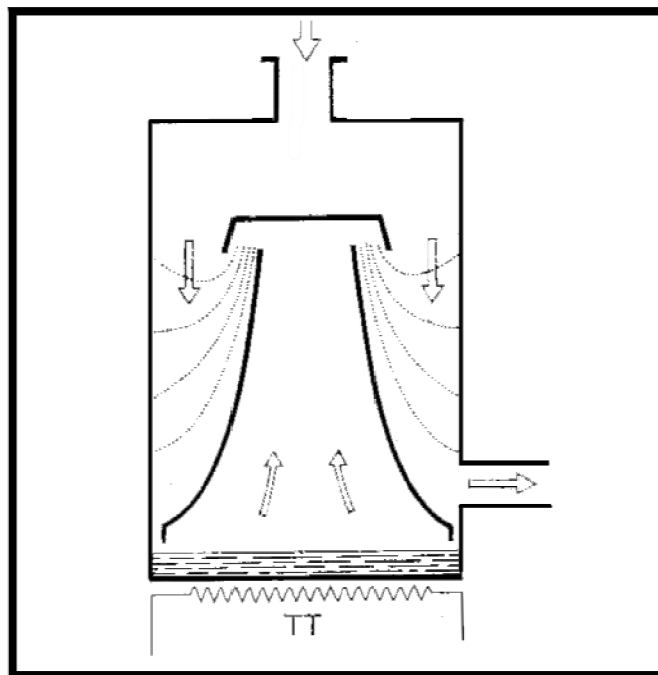
Modern vacuum technology allows to achieve pressure about 10^{-7} up to 10^{-10} Pa in chambers. For this scale of pressures, we cannot use universal vacuum pump, which allow such reduction of pressure. From physical point, vacuum technology offers us wide scale of vacuum pumps. If we want to reach, it is necessary to remove gases from capacity of system and from surface of system.

Rotary oil vacuum pump

Rotary oil vacuum pump is mechanical pump with a rotor with different shapes, which is gasketed and is lubricated with oil. There is part of transportation vacuum pump group, in which the gas is transported from entering to exit point. Pumping effect is generated by periodical shrinking and enlarging of working area evoked by rotation of rotor.

Diffusion vacuum pump

Diffusion vacuum pump works on principle of interaction of chaotically gas with univergent bunch of pumped material. In Diffusion pumps the heated gases of pumped material stream through stream piping, which is situated in the middle of chest, to injector. The flow of the steam in injection changed orientation and head towards the bottom. Average free movement molecules of gasses is enough big to get deep into steam flow so probability of return is low.

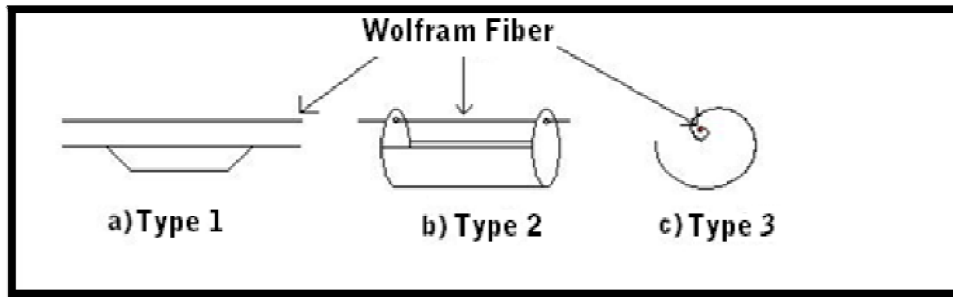


Picture 10 - Model of diffusion vacuum pump

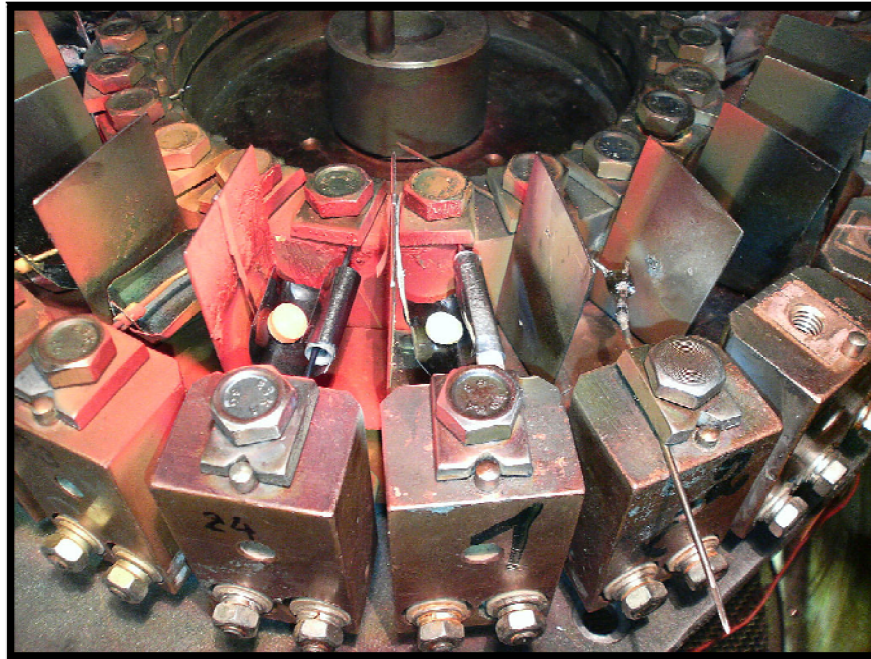
Evaporator

Indirect heated evaporators were used in order to avoid material melting and droplets spitting. Material was heated and evaporated from its surface by heat emission from the tungsten filament. The boat depicted in fig 12c was the most efficient due to radiation focusing effect.

Evaporator is carrier of evaporated material, which is heated and then evaporated



Picture 11 - Models of different kind of evaporators

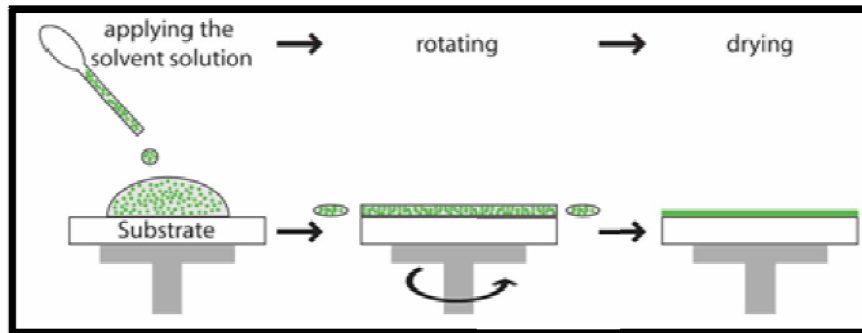


Picture 12 - Evaporators

The most effective type is type 3 where the losses generated by radiation are minimal.

Spin coating

Spin coating is cheap and fast method to creating a homogeneous thin films on flat substrates. A small amount of solution is spread on substrate, which is fixed and high speed rotating to spread the solution. The results of spin coating depend on viscosity of solution, speed of rotation and time of rotation.



Picture 13 - Spin coating process

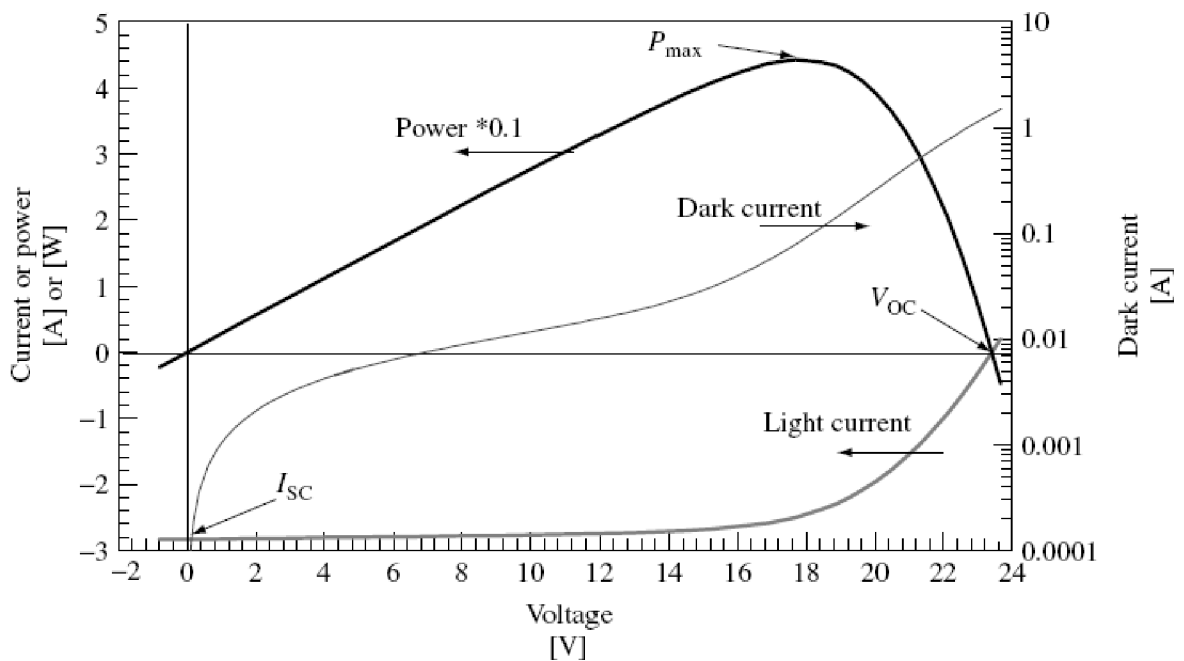
Electrical measuring

There are two main characteristics, which describe the performance of solar cells, current versus voltage and spectral response versus wavelength.

Current versus voltage measurement

A solar cell can be modeled as a diode in parallel with current generator[1]. One of the most important parameter we can calculate from VI characteristic is fill factor. The fill factor is a normalized parameter indicating how ideal the diode properties are and it is calculated by the following expression:

$$FF = \frac{P_{max}}{V_{oc}I_{sc}} \quad (3)$$



Picture 14 - Current versus voltage diagram

Spectral responsibility

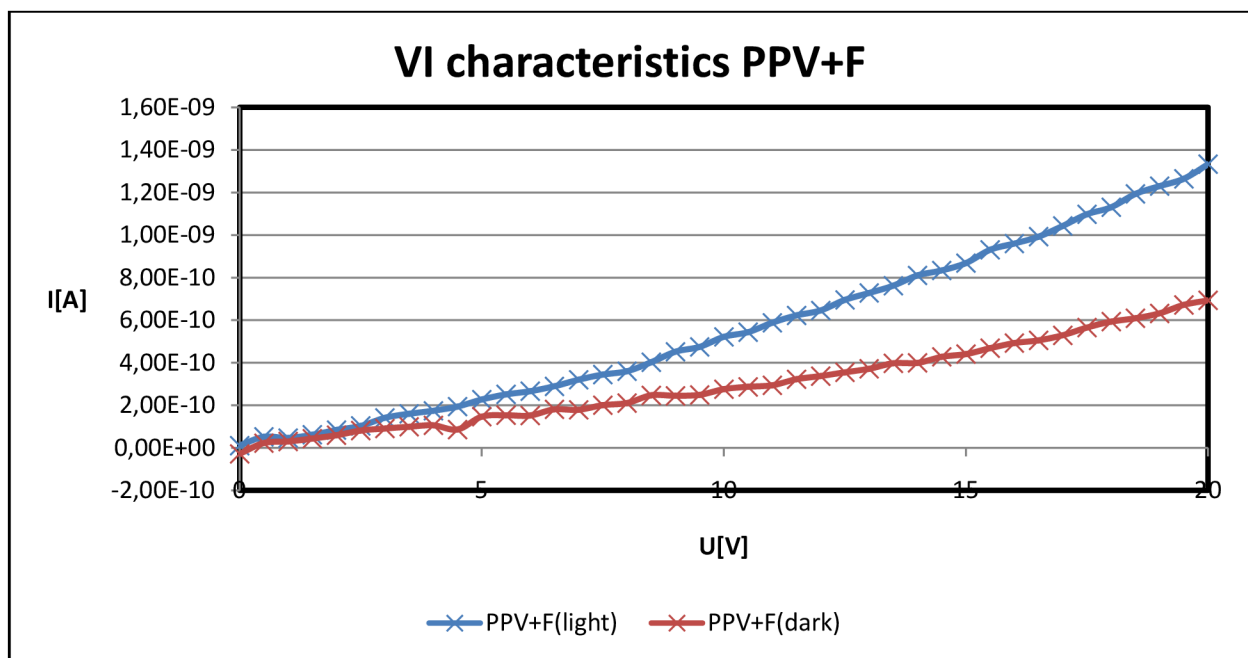
Spectral responsibility measuring can tell us a lot about generation, recombination and diffusion in photovoltaic devices. The spectral response is measured in units of current

produced per unit power or electron – hole pairs produced per incident photon through the equation:

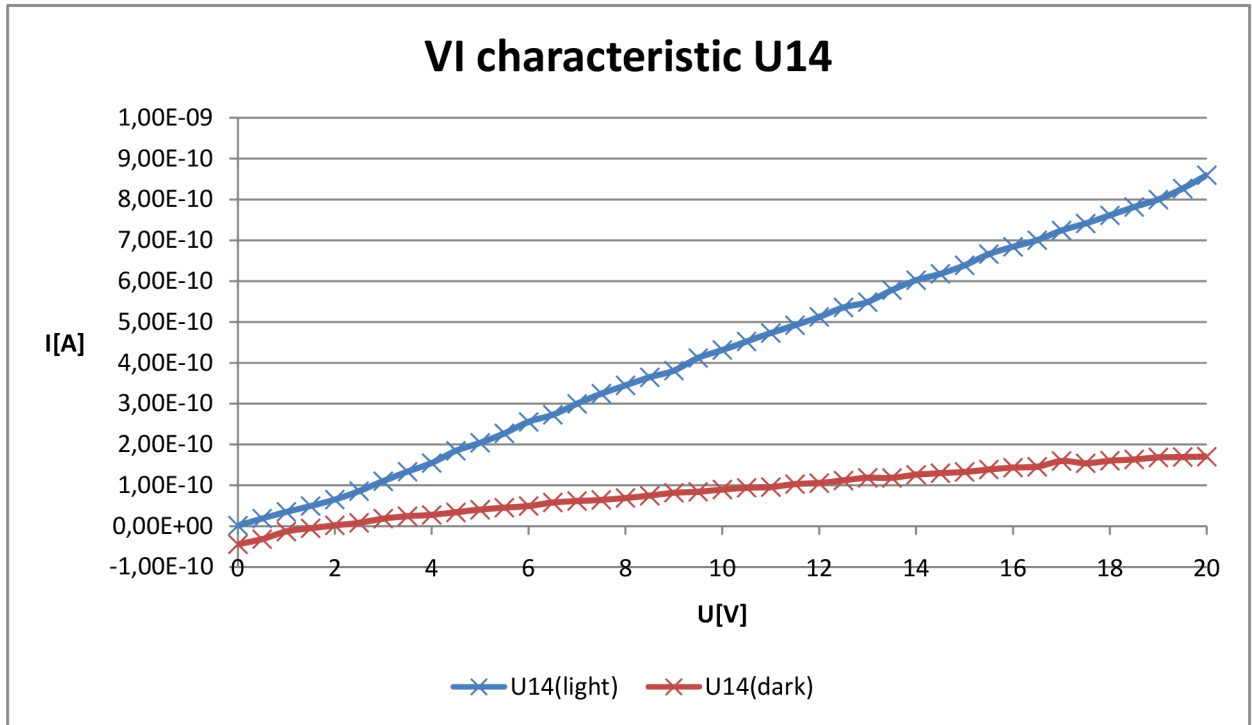
$$QE(\lambda) = \frac{qS(\lambda)}{\lambda hc} \quad (4)$$

All electrical measurements were made on the Faculty of Chemistry, Brno University of Technology, at automated workstation. Workstation consists of personal computer, voltage source and ampere-meter. All hardware is controlled through software interface prepared in Labview from National Instruments. All materials were measured in two different conditions. Firstly, we measured the VI characteristics in dark conditions and the second measurement was made with sample exposure to the light from the halogen lamp. This shows us if the conductivity depends on illumination.

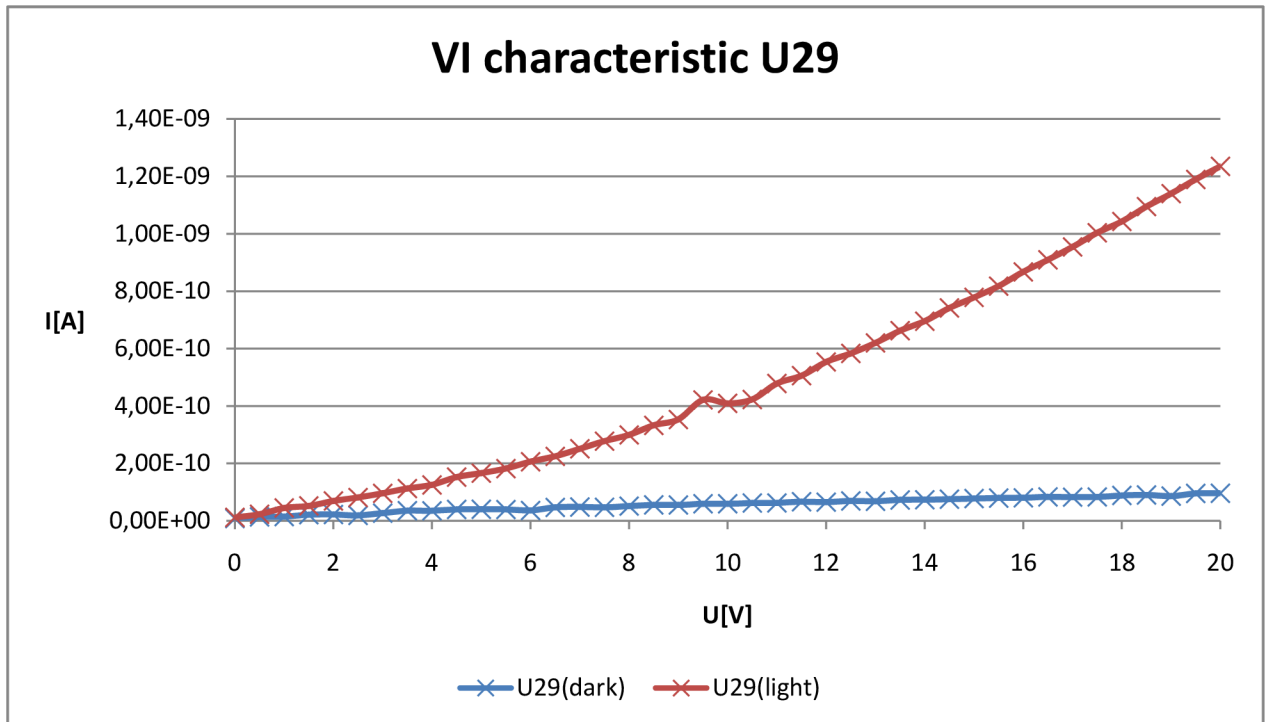
As a reference material, we decided to use a PPV+F, which we prepared with spin coating technology. For material U35 with had a two samples with different thickness.



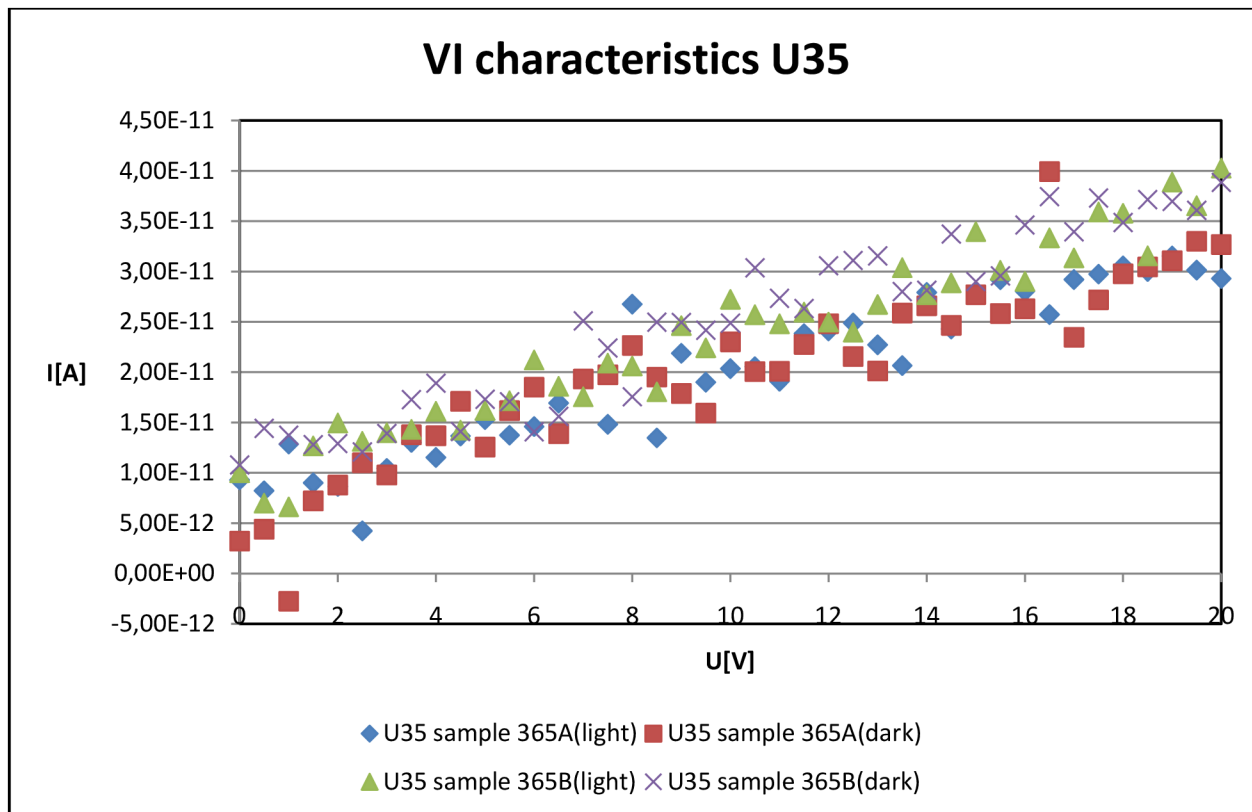
Picture 15 - VI characteristics PPV+F



Picture 16 - VI characteristics U14



Picture 17 - VI characteristics U29



Picture 18 - VI characteristics U35

From the results, we can see that the materials U29 and U14 are photoconductive and the conductivity of material U35 does not depend on illumination.

Conclusion

In the beginning of the work are examined the effects, which happened in organic semiconductors. The band theory, which helps us explain effect in inorganic semiconductors should, helps us, it could help us explain some effects in organic, because the structure of organic semiconductors is more complicated.

In the next part of the work is examined of the influence of different structures on the performance of the organic solar cells. Single layer structure does not have very good efficiency, but it should be very useful for examination for measuring of attributes of used organic material. If it is used two different materials it should be created double layer structure, which should combine advantages of both materials, but the thickness of layers increase and negative effects should appear. This problem should be solve the blend structure, which is created by mixture of two materials. The most preferred structure for organic solar cells is sandwich structure, which has the best efficiency and allows us to use two different technologies for preparing the cells.

In experimental part is examined the attributes materials, which were available. The samples with materials U14 and U29 appears increase of current under illumination in comparison with measurement in dark, so this materials are suitable for further research. The samples with material U35 we found out that the current does not depend on the illumination, so we should not count with him in usage for solar cells.

From the results, it is clear that some organic materials are photosensitive, but for the choice of the most suitable material is it necessary to do more research. It is necessary to enlarge amount of measured materials and measure their VI characteristics, spectral responsibility, influence of different structures and eventually the influence of different dopants on conductivity.

For the future, we should forecast that organic materials should reach sufficient efficiency to be used in commercial applications and be the competitor to inorganic solar cells

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Used shortcuts, symbols and constants

A	acceptor
D	donor
I	electric current
M	molecular weight
p	preassure
t	temperature
U	voltage