Materiais Nanoestruturados e sua aplicação na área de conversão de energia solar

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www.lnes.iqm.unicamp.br

UNICAMP

Outline

- Breve Introdução sobre energia fotovoltaica
- Nanomateriais e Energia Solar
- Introdução às Células Solares de Terceira Geração (3G)
- Uso de nanomateriais em células de TiO_2/corante focando em:
  - Nanobastões e nanotubos de óxidos semicondutores. Compósitos com nanotubos de carbono
- Uso de nanomateriais em células orgânicas focando em:
  - Uso de nanopartículas de calcogenetos
  - Nanotubos de Carbono, Grafeno e seus compósitos

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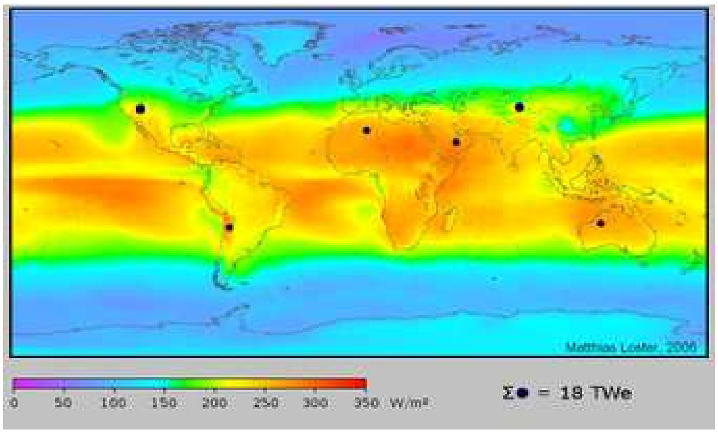
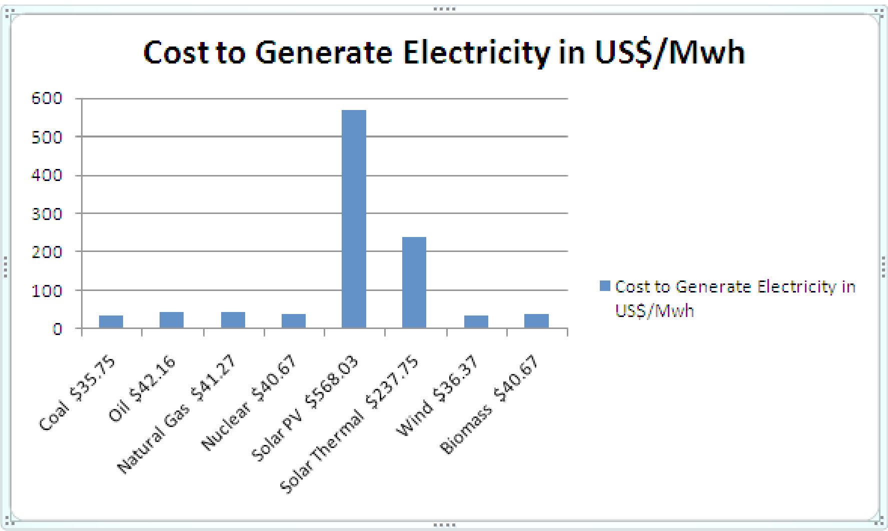
Energia Solar

Pontos em preto: área necessária para suprir a demanda de energia

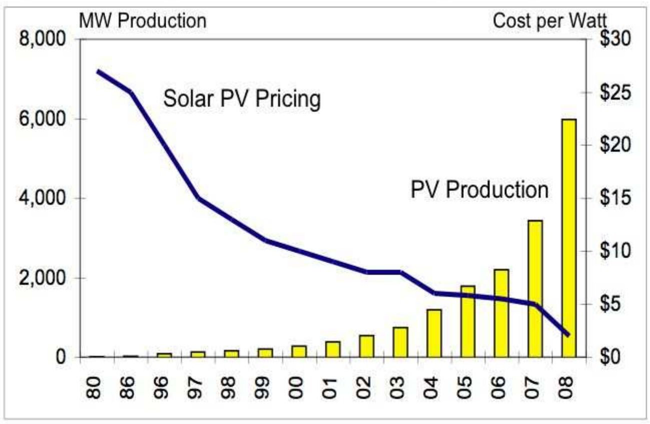


❖ Em apenas 1 hora o Sol despeja na Terra uma quantidade de energia superior ao consumo global de um ano inteiro!!!

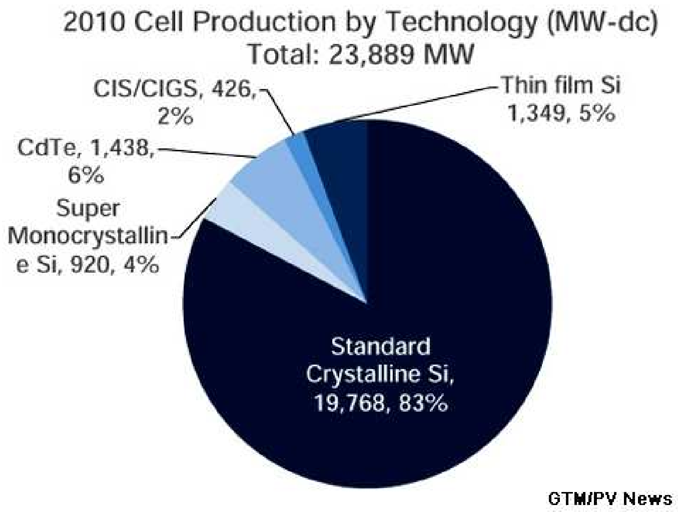


https://matteranenergy.us/Joe%20Sixpack's%20Technology%20Page.html

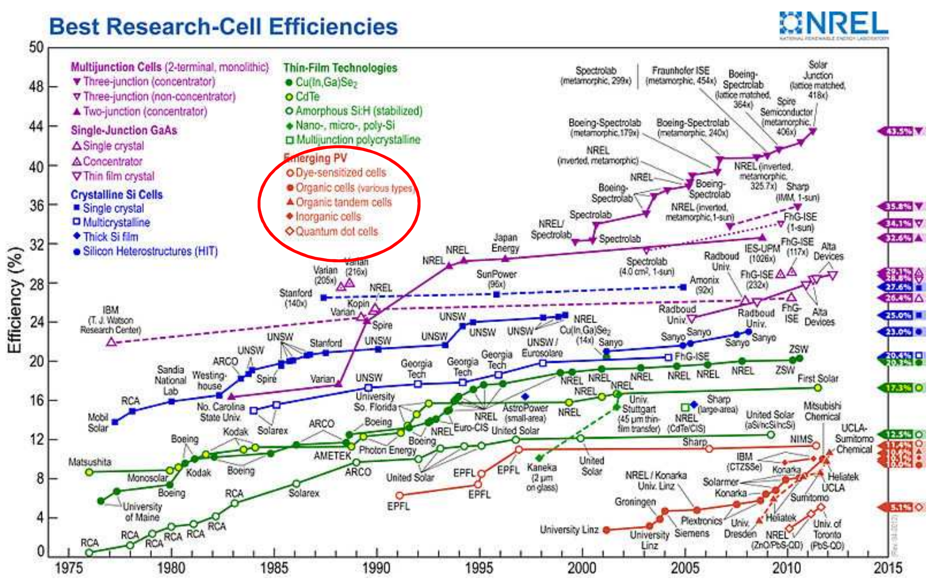
Solar PV Global Production and Cost per Watt



Source Solar Buzz, Company reports,, Green Econometrics research



http://imaginesolar.com/news/articles-on-imaginesolar/



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How the nanotechnology can help in solar energy conversion?

◇ Cost
◇ Efficiency
◇ Flexibility

Nanotechnology and Solar Cells

- Improved efficiencies: novel nanomaterials can harness more of the sun's energy; can transport charges more efficiently;
- Lower costs: some novel nanomaterials can be made cheaper than alternatives;
- Reduced manufacturing costs as a result of using a low temperature process (e.g. printing) instead of the high temperature vacuum deposition;
- Flexibility: traditional silicon based solar cells are brittle, nanotechnology can develop devices that can be flexible, so that entire buildings could be covered.

Nanomaterials and Solar Cells

- Nanomaterials are those which have structured components with at least one dimension between approximately 1 and 100 nm
- Two principal factors cause the properties of nanomaterials to be different from the bulk materials:
  - Increased relative surface area
  - Quantum effects

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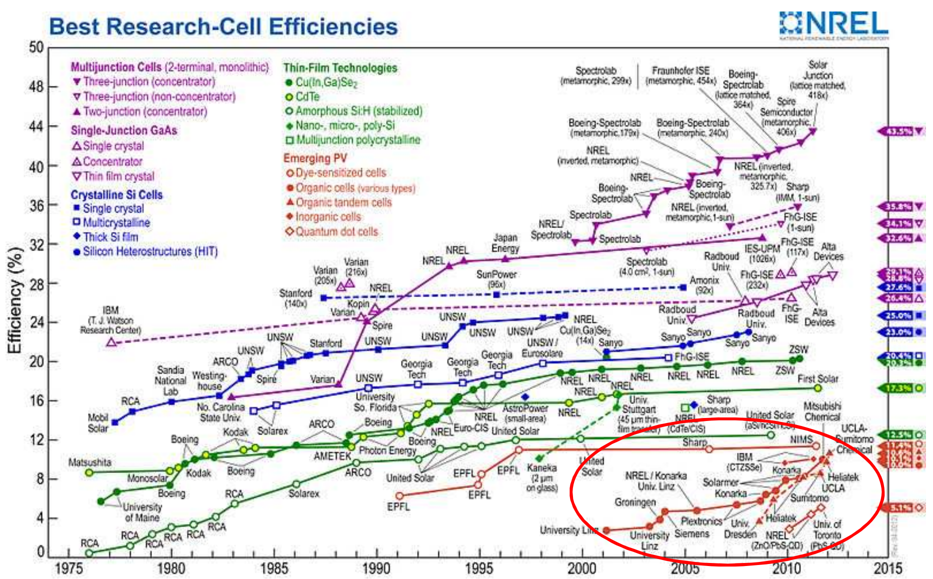


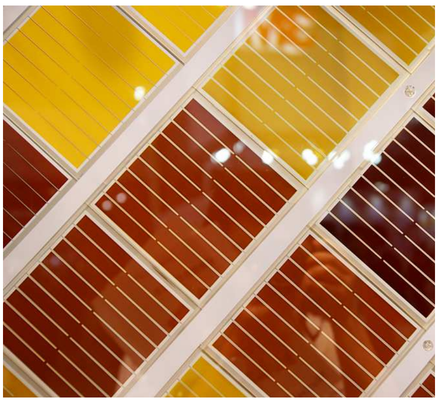
Dye Sensitized and Organic Solar Cells (*3G solar cells*): common features

- Low cost: materials and processes are cheap
- Ink jet, spin coating, roll-to-roll

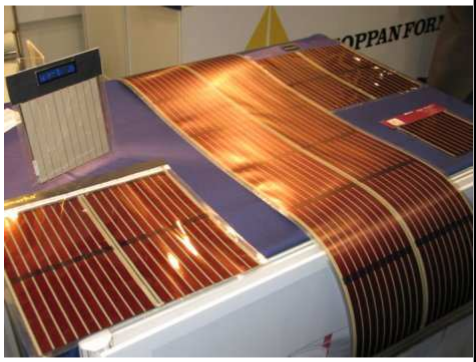
- Low temperature
- Colored
- Flexible
- Semitransparent

- Diverse types or organic and inorganic materials
- Optical and transport properties can be easily tuned

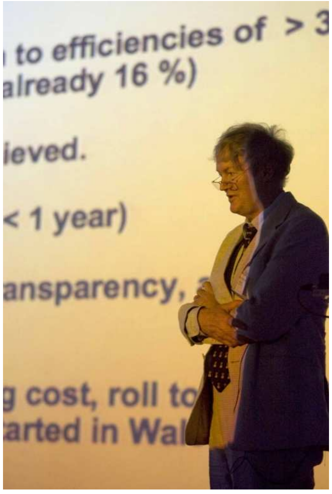
3G Solar Cells (or NanoPV)



TiO_2/Dye Sensitized Solar Cells - DSSC



Organic (or Hybrid) Solar Cells



Prof. Michael Gratzel

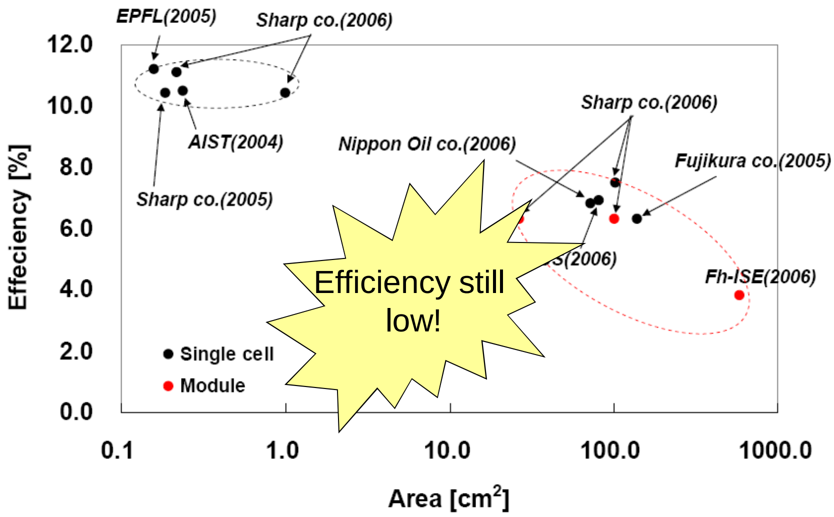
Prof. Alan Heeger –Chemistry Nobel Prize in 2000

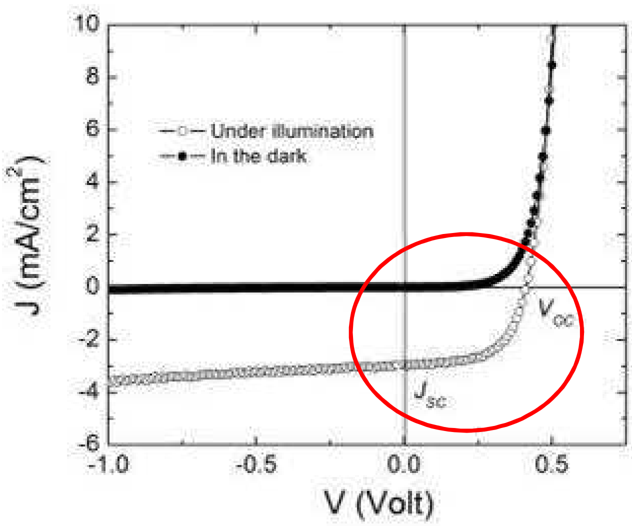
Dye Sensitized and Organic Solar Cells: applications

www.g24i.com

27.06.12 - Dye-sensitized solar cells (DSSC) from EPFL enter the public market. Logitech chose this technology to power its new flagship product

Dye Sensitized and Organic Solar Cells: disadvantages

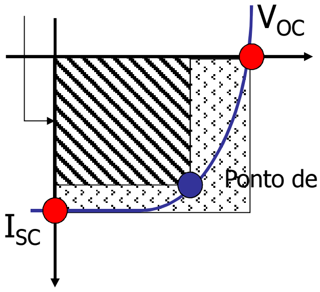




A célula solar opera no quarto quadrante!

Caracterização das células solares

Fator de preenchimento (FF)



$$FF = \frac{P_{max}}{P_{th}} = \frac{I_m \times V_m}{I_{SC} \times V_{OC}}$$

$$\eta = \frac{I_{SC} \times V_{OC} \times FF}{100^*}$$

* 100 mW cm^{-2} ou 1000 Wm^{-2}

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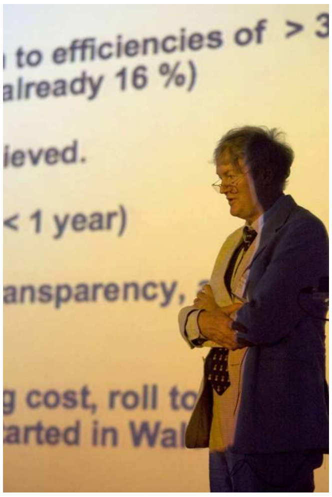
Nanomaterials in Dye Sensitized Solar Cells

Nanomaterials in Gratzel's cells



A low-cost, high-efficiency solar cell based on dye-sensitized colloidal TiO_2 films

Brian O'Regan* & Michael Grätzel†

Institute of Physical Chemistry, Swiss Federal Institute of Technology, CH-1015 Lausanne, Switzerland

THE large-scale use of photovoltaic devices for electricity generation is prohibitively expensive at present: generation from existing commercial devices costs about ten times more than conventional methods[1]. Here we describe a photovoltaic cell, created from low- to medium-purity materials through low-cost processes, which exhibits a commercially realistic energy-conversion efficiency. The device is based on a 10-μm-thick, optically transparent film of titanium dioxide particles a few nanometres in size, coated with a monolayer of a charge-transfer dye to sensitize the film for light harvesting. Because of the high surface area of the semiconductor film and the ideal spectral characteristics of the dye, the device harvests a high proportion of the incident solar energy flux (46%) and shows exceptionally high efficiencies for the conversion of incident photons to electrical current (more than 80%). The overall light-to-electric energy conversion yield is 7.1–7.9% in simulated solar light and 12% in diffuse daylight. The large current densities (greater than 12 mA cm^{-2}) and exceptional stability (sustaining at least five million turnovers without decomposition), as well as the low cost, make practical applications feasible.

Nature, 1991

Dye Sensitized Solar Cells or Gratzel's cells

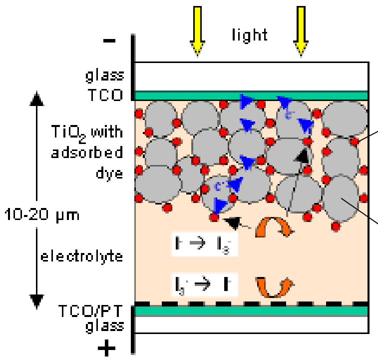
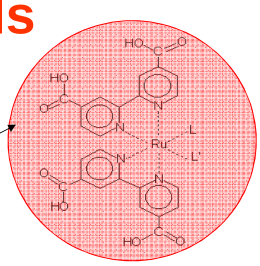
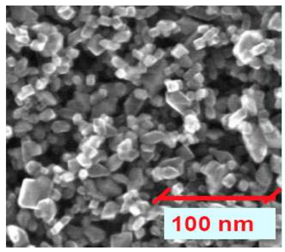


Fig. 1: Schema of dye-sensitezed solar cell





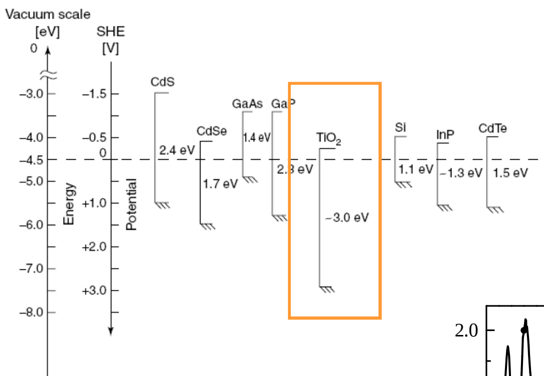
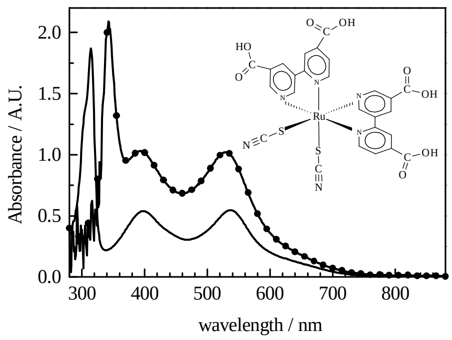
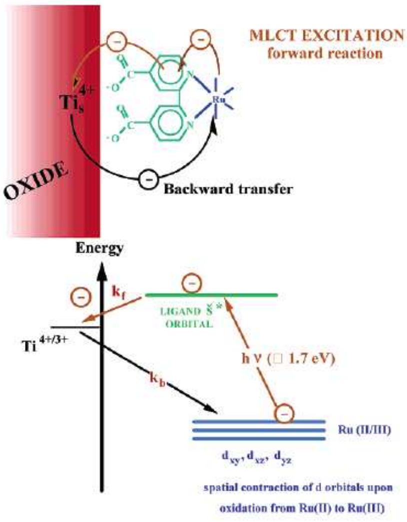


Fig. 8 Relative dispositions of various semiconductor band edge positions shown both on the vacuum scale and with respect to the SHE reference. These band edge positions are for an aqueous medium of pH ~1.



The electron transfer process...



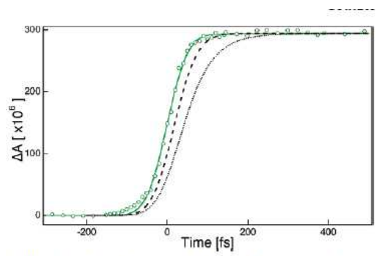
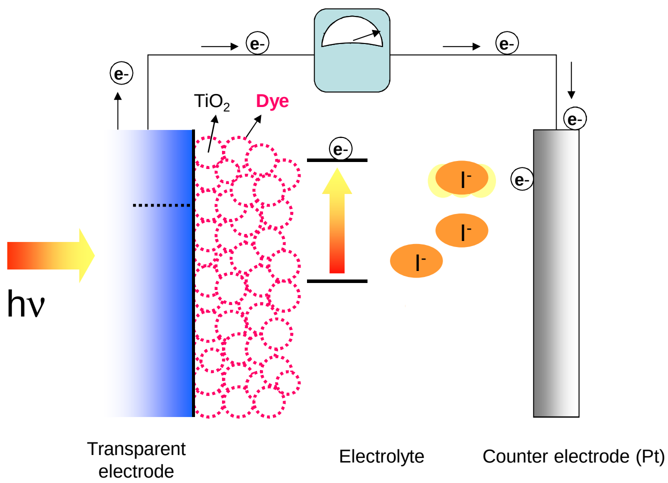


Figure 7. Transient absorption signal for N-719 adsorbed on nanocrystalline titania (○) (pump wavelength 535 nm; probe 860 nm). The fitted instrument response is 57 fs (—). The simulated exponential rises with time constants of 20 fs (– –) and 50 fs (···) and convolved with the same instrument response are shown.

Inorg. Chem. **2005**, *44*, 6841–6851

Inorganic Chemistry Article



Recombination

$e^-(TiO_2)CB + TiO_2/Dye^+ \longrightarrow TiO_2/Dye$

$e^-(TiO_2)CB + 1/2\ I_3^- \longrightarrow 3/2\ I^-$

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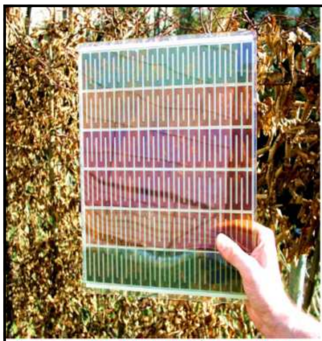
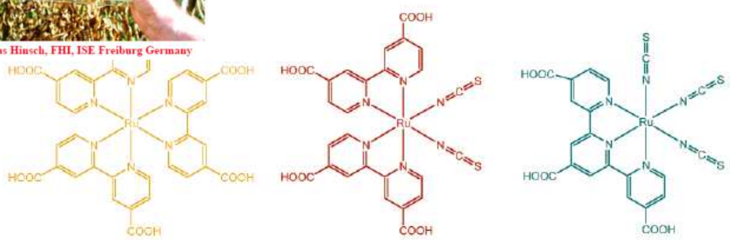
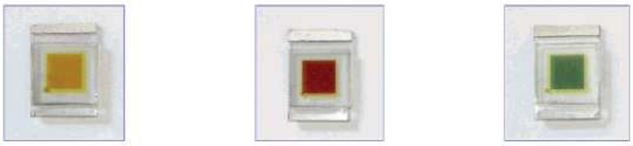
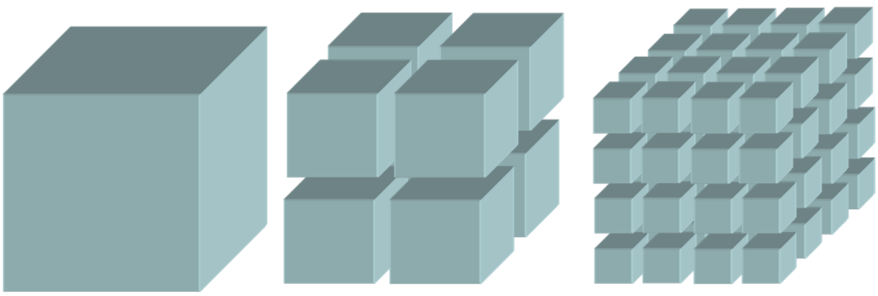


Courtesy Dr. Andreas Hinsch, FHI, ISE Freiburg Germany





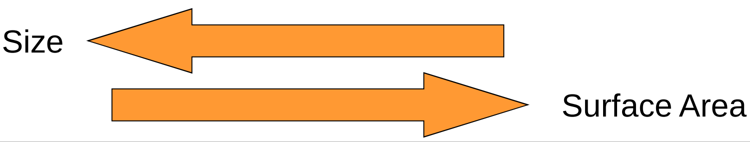
Gratzel's cells are *per si* an example nanomaterials applications



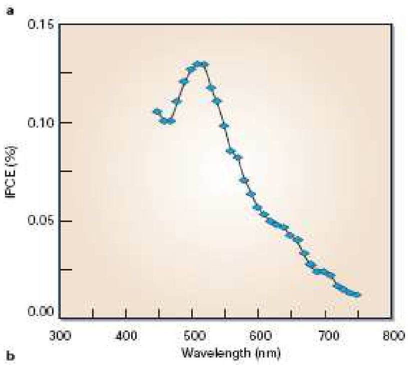
1 cubo de lado *m*
Volume: m^3
Área superficial: 6 x m^2

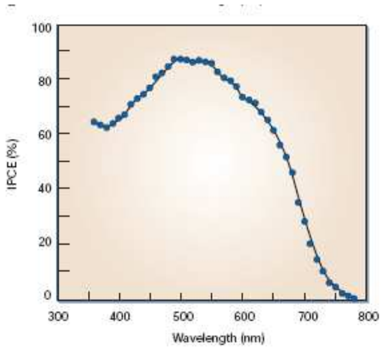
8 cubos de lado *m/2*
Volume: m^3
Área superficial: 12 x m^2

64 cubos de lado *m/4*
Volume: m^3
Área superficial: 24 x m^2



IPCE ~ eletrons/photons





b

Figure 5 The nanocrystalline effect in dye-sensitized solar cells. In both cases, TiO_2 electrodes are sensitized by the surface-anchored ruthenium complex *cis*-$RuL_2(SCN)_2$. The incident-photon-to-current conversion efficiency is plotted as a function of the excitation wavelength. **a**, Single-crystal anatase cut in the (101) plane. **b**, Nanocrystalline anatase film. The electrolyte consisted of a solution of 0.3M LiI and 0.03M I_2 in acetonitrile.

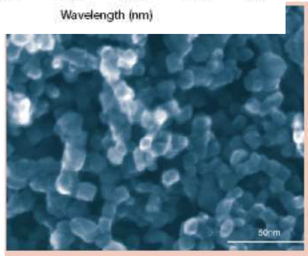
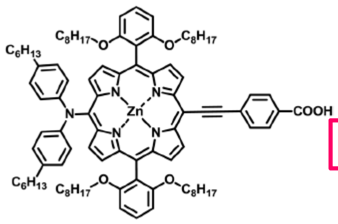


Figure 4 Scanning electron micrograph of the surface of a mesoporous anatase film prepared from a hydrothermally processed TiO_2 colloid. The exposed surface planes have mainly (101) orientation.

Michael Grätzel
NATURE | VOL 414 | 15 NOVEMBER 2001 Photoelectrochemical cells

Champion Gratzel's cell

4 NOVEMBER 2011 VOL 334 **SCIENCE**

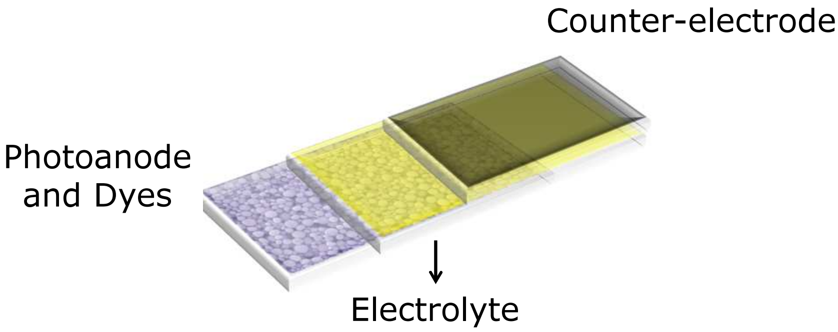


at 995 W/m^2 intensity. The cosensitization of **YD2-*o*-C8** with the previously prepared organic D-π-A dye, coded **Y123**, yielded an efficiency of 12.3% when used in conjunction with the $Co^{(II/III)}$tris(bipyridyl)–based redox electrolyte. The PCE even exceeds 13% under AM 1.5 solar light of 500 Wm^{-2} intensity.

Table 1. Detailed photovoltaic parameters of the devices made with the dyes **YD2** and **YD2-*o*-C8** and cobalt-based AY1 electrolyte at different light intensities. P_{in}, incident intensity of AM1.5 solar light.

Dye	Electrolyte	P_{in} (mW/cm^2)	J_{sc} (mA/cm^2)	V_{oc} (mV)	FF	PCE (%)
YD2	AY1	9.4	1.5	745	0.82	9.5
		51.3	8.0	805	0.76	9.5
		99.8	14.9	825	0.69	8.4
YD2-*o*-C8	AY1	9.4	1.7	875	0.77	12.5
		51.2	9.3	940	0.74	12.7
		99.5	17.3	965	0.71	11.9

LNES Activities in DSSC

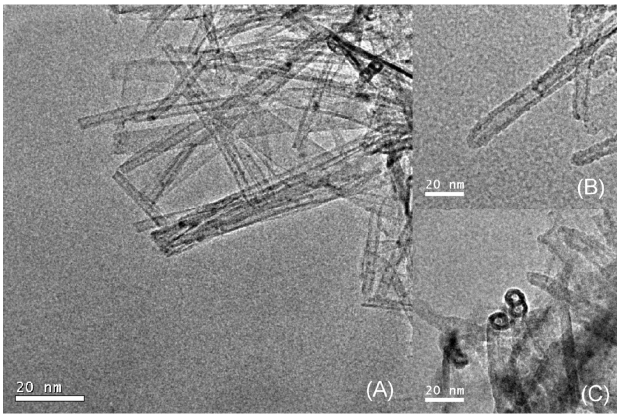


□For review: Coord. Chem. Rev. 2004 and J. Mater. Chem 2009

Nanorods, Nanotubes, Nanowires and Nanoflowers in Dye Cells

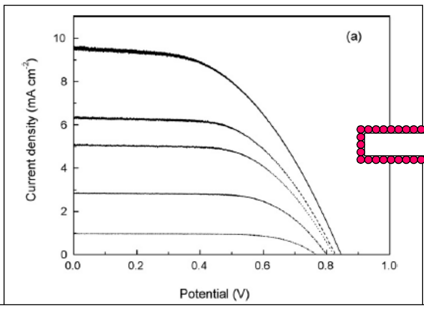
- These nanostructures can increase solar cell's efficiency by;
  - Increasing the electron transport and minimizing recombination
  - Harvesting more light due to an increase in the surface area (more dye molecules can be adsorbed)
  - Harvesting more light due to light scattering
  - Allowing a better penetration of a polymer or gel electrolyte

◇ TiO_2 nanotubes, Nogueira and cols. J. Photochem. Photobiol., 2006



ϕ ~3 nm
L = 100 nm -1 μm

- Growth by hydrothermal method



❖ NaI + (P(EO-EPI)$_{84:16}$ + 40 wt % P

- TiO_2 nanotube films allow more dyes to be chemically absorbed (high surface area)
- Its open structure increases ionic diffusion
- 1D structure increases electron transport

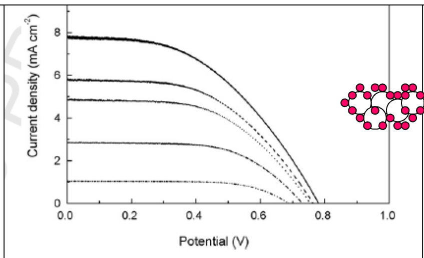


Fig. 4. Comparison of the current–potential curves obtained under different light intensities for the TiO_2 dye-sensitized solar cell assembled with the plasticized polymer electrolyte P(EO-EPI)84:16/PEG-diB + 12 wt% NaI/I_2: (a) device with TiO_2 nanotube film and (b) device with nanoparticulated film.

Table 1
Comparison of the electrical characteristics of the dye-sensitized solar cells investigated in this work

I_{RR} (mW cm^{-2})	I_{SC} (mA cm^{-2})	V_{OC} (V)	FF (%)	η (%)
DSSC assembled with TiO_2 nanotubes and plasticized polymer electrolyte				
100	9.63	0.846	49	4.03
9	0.97	0.767	71	5.91
DSSC assembled with TiO_2 nanoparticles and plasticized polymer electrolyte				
100	7.75	0.781	47	2.84
9	1.04	0.688	66	5.28

◇ TiO_2 fibers, J. Mater. Chem., 2010

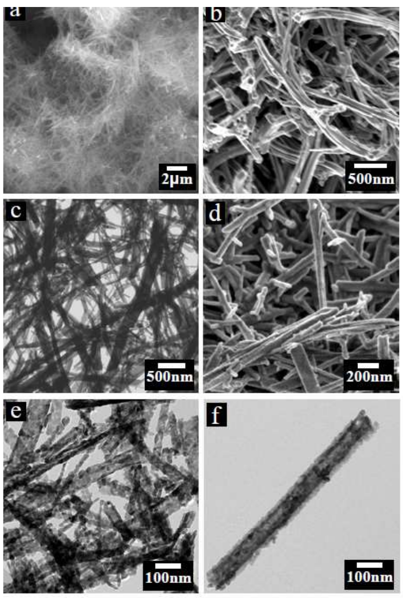


Figure 5. SEM (a and b) and TEM (c) images of as-synthesized titanium glycerolate fibers, and SEM (d) and TEM (e - f) of the anatase nanorods.

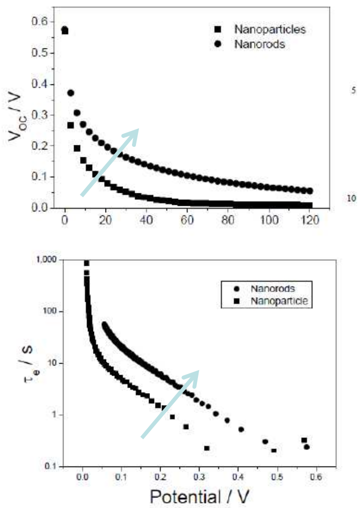
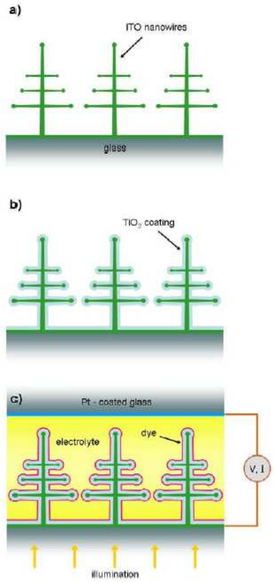


Figure 8. (a) Decay of V_{oc} as a function to time for DSSCs prepared with TiO2 nanoparticles and nanorods. (b) Electron lifetimes for the aforementioned two cells as a function of the open circuit potential.

◇ ITO nanowires, Nogueira and cols. Scripta Mater., 2007



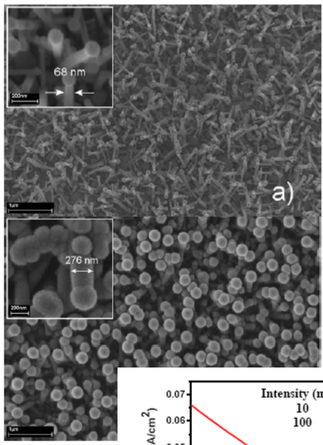


Figure 2. FE-SEM image deposition of the TiO_2 lay thickness measurements.

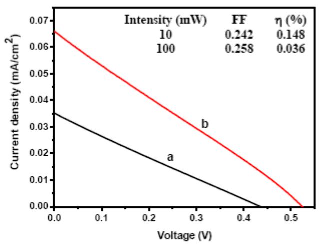
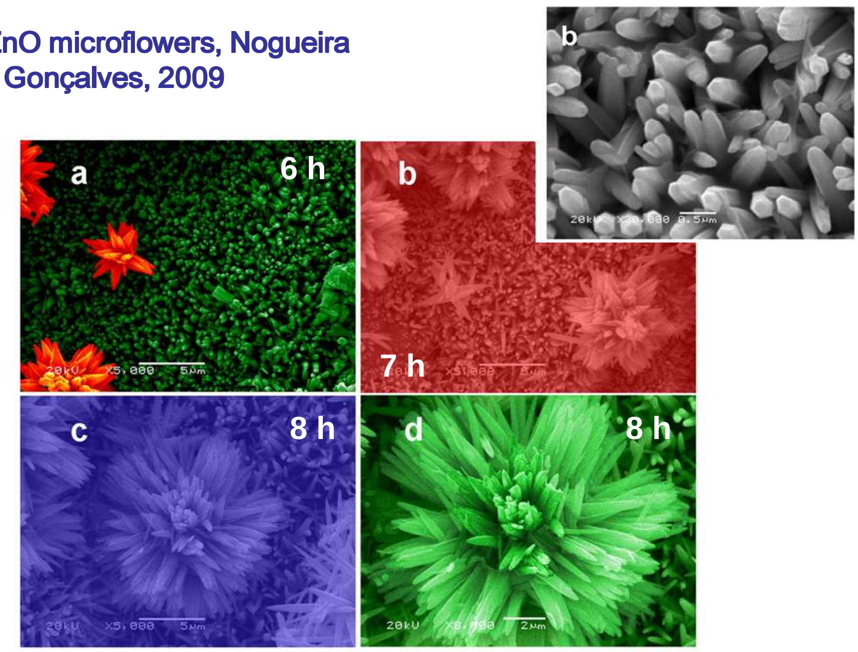
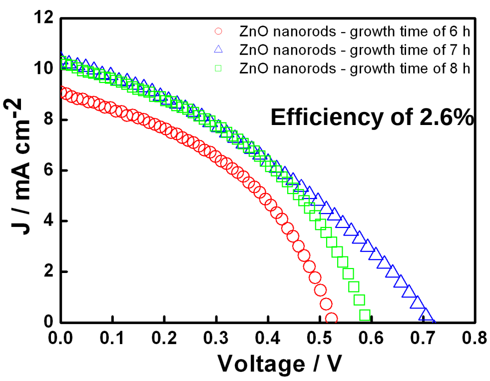


Figure 3. I-V curves for the 3D DSSC assembled with a brush-like TCO electrode under different light intensities: (a) 100 mW cm^{-2} and (b) 10 mW cm^{-2}.

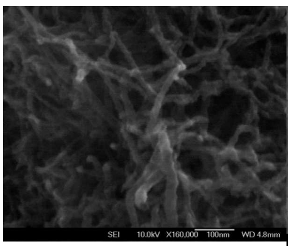
◇ ZnO microflowers, Nogueira and Gonçalves, 2009



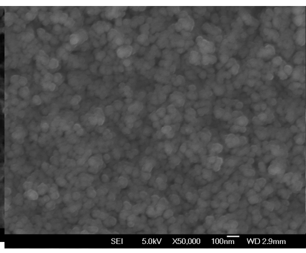


- The increase in the photocurrent is attributed to an increase in dye loading and scattering effects

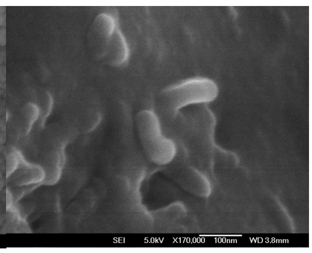
Inorganic nanocomposites with carbon based materials



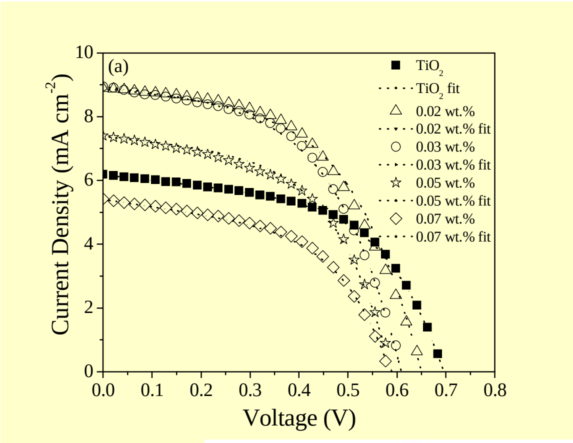
MWCNT

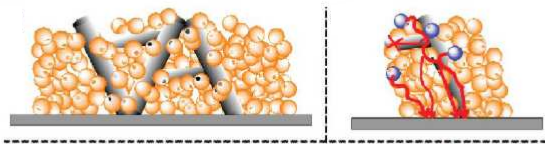


NC-TiO_2



Composite





- Andreia de Morais et. al., J. Photochem. Photobiol, in press.

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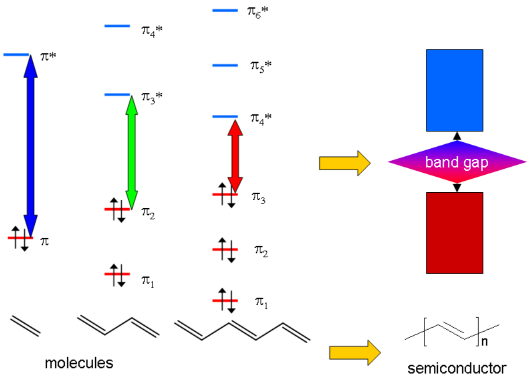
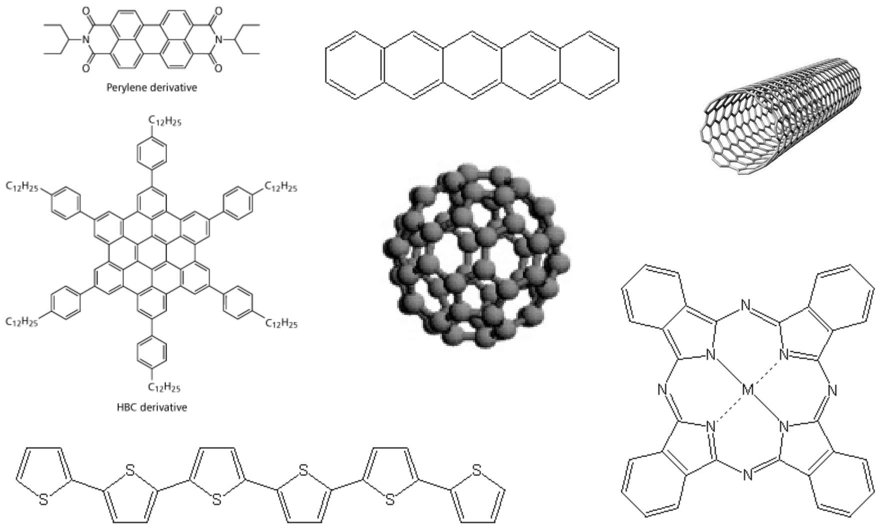
Nanomaterials in Organic Solar Cells

Organic Semiconductors

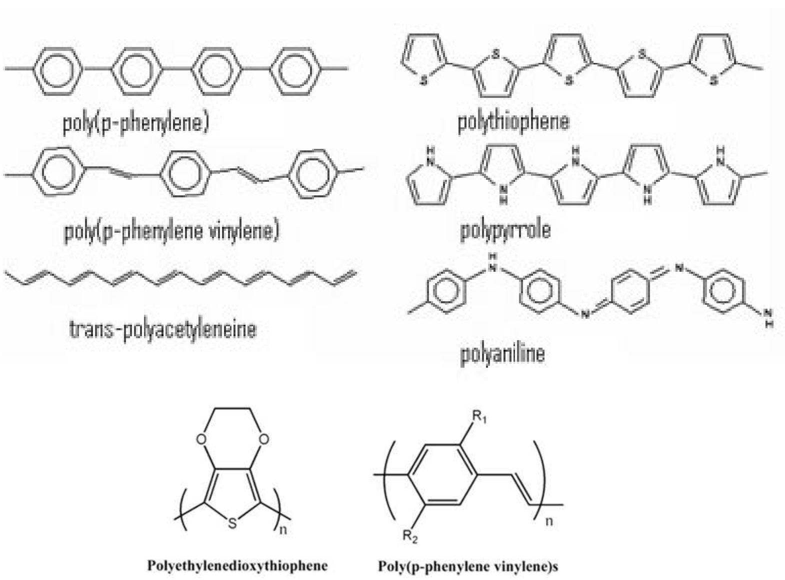
Materiais conjugados onde tanto a absorção ótica quanto o transporte de carga são dominados pelos orbitais moleculares π e π* parcialmente deslocalizados



Organic Semiconductors



Conducting polymers



The Royal Swedish Academy of Sciences has awarded the Nobel Prize in Chemistry for 2000 jointly to **Alan J. Heeger, Alan G. MacDiarmid** and **Hideki Shirakawa** "for the discovery and development of conductive polymers".

Alan G. MacDiarmid
Professor at the University of Pennsylvania, Philadelphia, USA.

Hideki Shirakawa
Professor Emeritus, University of Tsukuba, Japan.

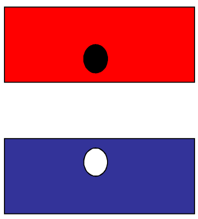
Alan J. Heeger
Professor at the University of California at Santa Barbara, USA.

Photoexcitation in organic semiconductors

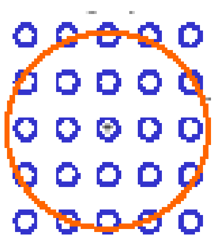
SC inorgânico

Banda de condução

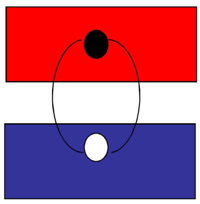
Banda de valência



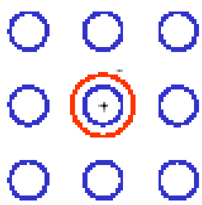
energia de "ligação"~ meV ➔
Elétron e buraco "livres"



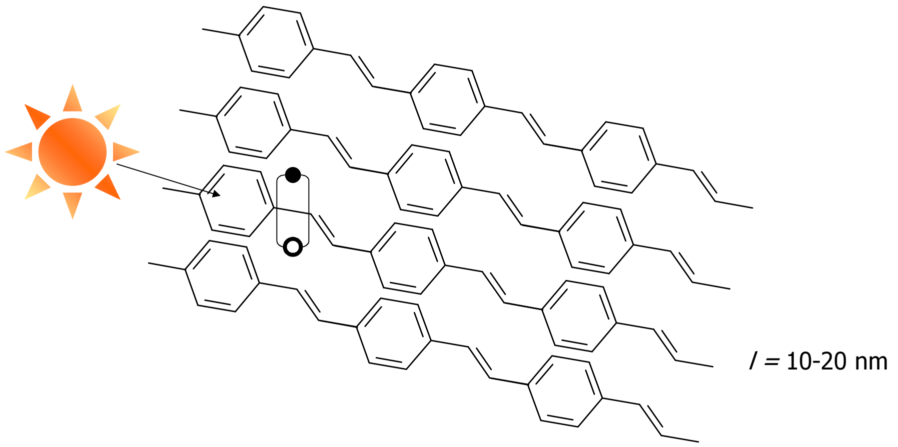
SC orgânico



energia de "ligação"
~ 0.4 a 1 eV ➔ ***excitons***

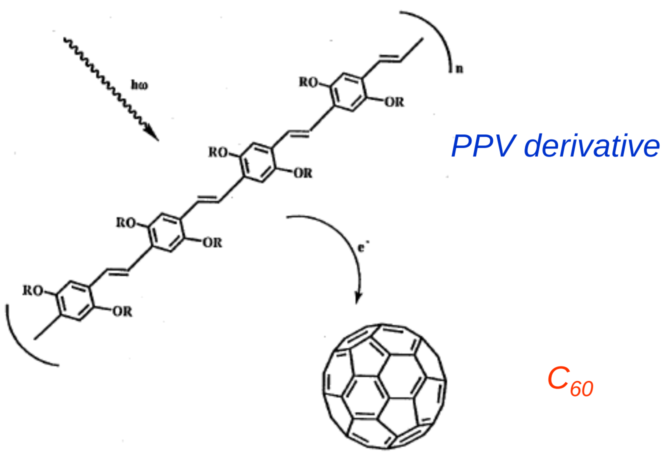


Photoexcitation in organic semiconductors



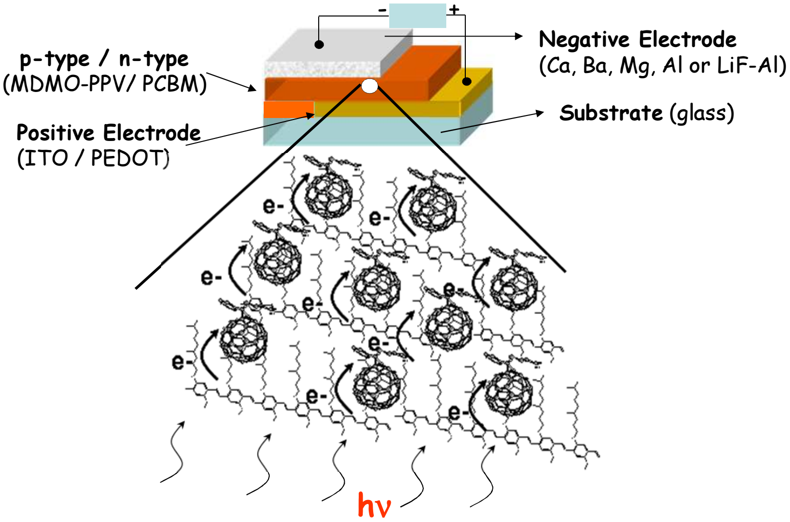
We need to separate the electron-hole pair in order to have free charges!

- Photoinduced electron transfer between a conducting polymer and C_{60} (~ 50 fs !!!)



- A. Heeger and co-workers, Science (1992)

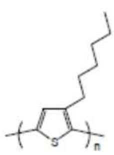
Bulk heterojunction solar cells



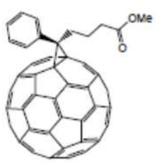
Efficiency 10% (Mitsubishi Co.)

- The bulk heterojunction is *per si* a nanostructured morphology!

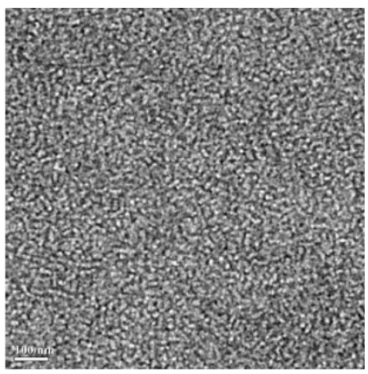
After 10 min annealing at 150 ^{o}C



P3HT



PCBM



Heeger, 2008

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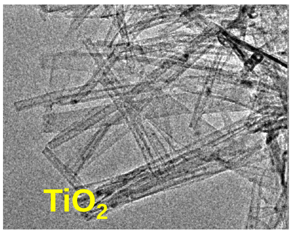
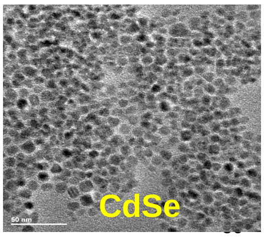
Nanoparticles in OSC: Hybrid SC

- Definition: When an inorganic material replaces one organic component in the “classical” organic solar cells (in most cases, the electron acceptor material)
  - Most used inorganic materials are nanoparticles:
    - TiO_2
    - ZnO
    - CdSe and CdS

o **Quantum confinement**
o **Large surface area**

Images from our group



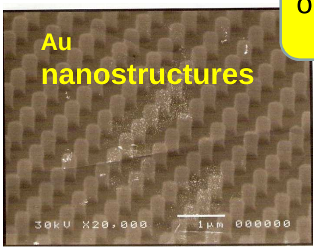


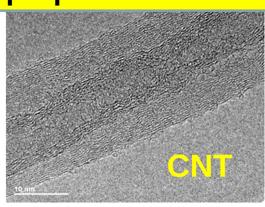
Nanoparticles in OPV: Hybrid SC

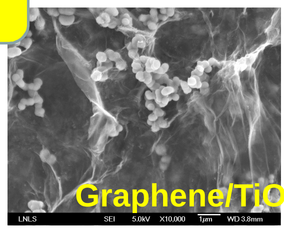
- Definition: When an inorganic material replaces one organic component in the “classical” organic solar cells (in most cases, the electron acceptor material)
    - Other materials are frequently used in hybrid devices
    - Gold and Silver nanostructures
    - Carbon based materials (carbon nanotubes and graphene)

- **Plasmon effects**
- **Improved transport properties**

Images from our group

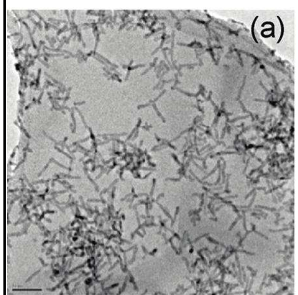


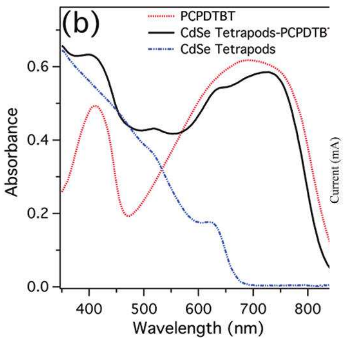


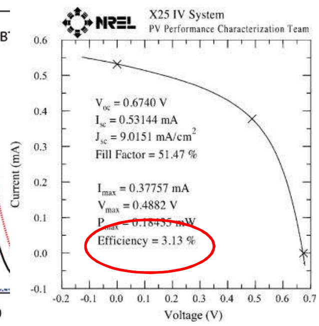


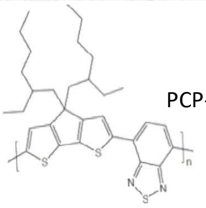
State of Art

HOPV based on CdSe or CdS









PCP-DTBT

S. Dayal et. al., Nano Letters, 10 (2010) 239

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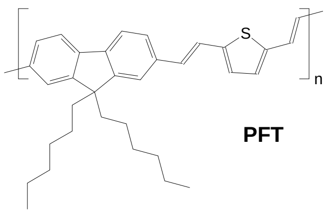
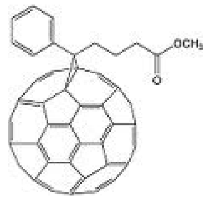
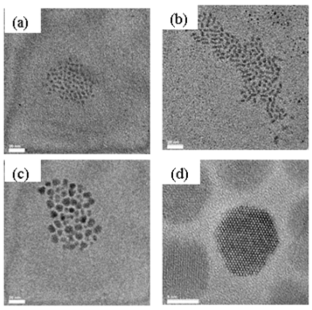
Introduction of CdSe nanoparticles in the mixture polymer-fullerene







- J. N. Freitas, I.R. Grova, L.C. Akcelrud, E.Arici, N. S. Sariciftci, A. F. Nogueira,
J. Mater. Chem., 2010, 20, 4845–4853
- J. N. Freitas, A. Pivrikas, B. F. Nowacki, L. C. Akcelrud, N. Serdar Sariciftci, A. F. Nogueira (cover sheet), Synth. Metals., 2010, 160, 1654-1661

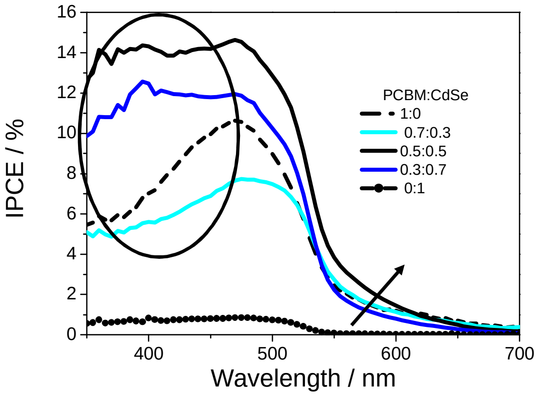
Introduction of CdSe nanoparticles in the mixture polymer-fullerene

- PFT + CdSe + PCBM
  - CdSe (4.0 nm) + PCBM = 80 wt %

PCBM:CdSe	Jsc (mA cm^{-2})	Voc (V)	FF (%)	η (%)
1:0	3.5	0.50	28	0.52
0.7:0.3	3.4	0.54	30	0.55
0.5:0.5	4.7	0.58	30	0.80
0.3:0.7	3.5	0.60	31	0.65
0:1	0.17	0.66	24	0.03

Optimum concentration of nanoparticles → The effect of the QD on the photocurrent can be due to light absorption AND/OR morphology

- IPCE (*incident photon to current effciency*)
  - IPCE = collected electrons / incident photons
    - As a function of wavelength



Slightly increase in the region 550-650 nm (CdSe absorption)

Change in IPCE profile in the region below 450 nm

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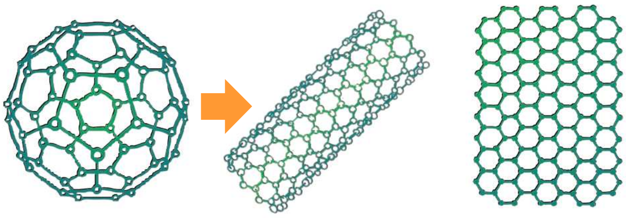
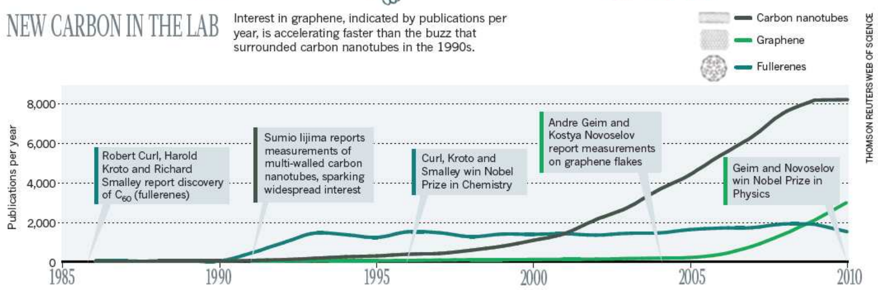
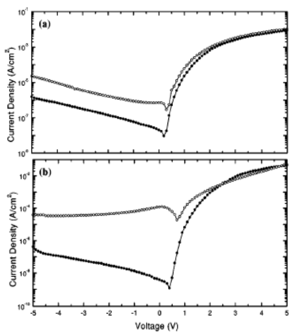
Carbon Nanotube and Graphene as Acceptor Materials

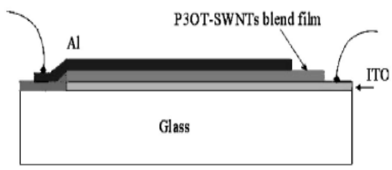




Carbon Nanotube and Graphene as Acceptor Materials

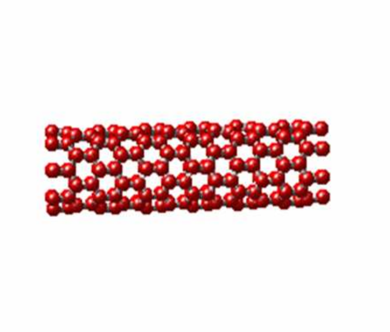
- Ago and co-workers (Phys. Rev. B, 2000): electronic interaction between PPV and MWCNT: energy transfer
- Kymakis and co-workers (Appl. Phys. Lett., 2001)



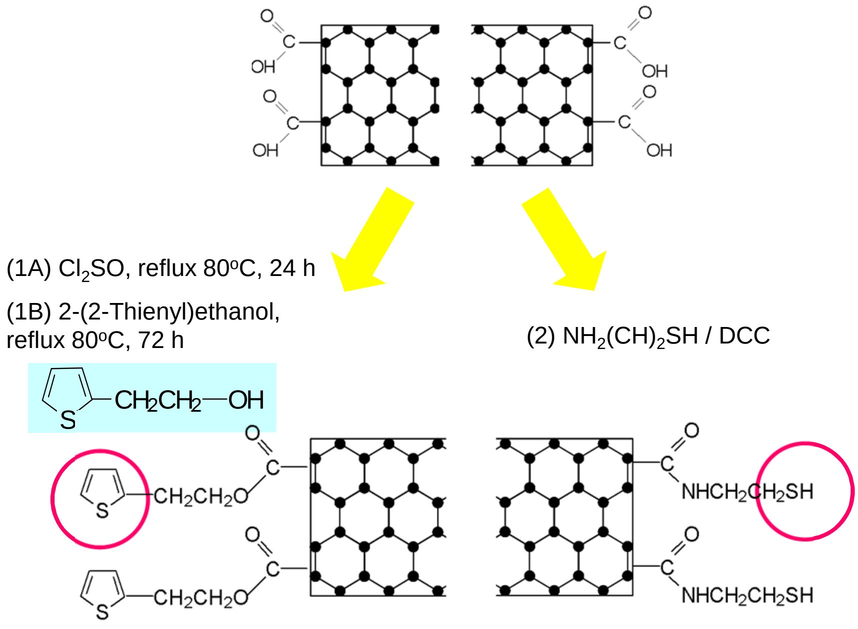


- V_{oc} = 0.7- 0.9 V
- I_{sc} = higher than the pristine polymer
- FF= 0,3 - 0,4

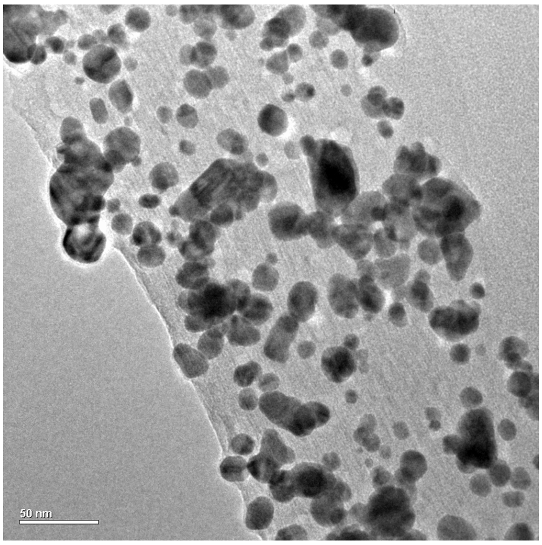
Carbon Nanotubes in OPV



- Excellent electronic, magnetic and mechanical properties
- Easy to functionalize
- High aspect ratio (length/diameter)
- **Metallic and Semiconducting states**
- **Polymer and CNT do not form homogenous dispersion**

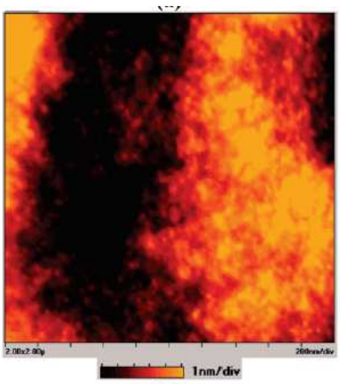


Nogueira and cols., J. Phys Chem C., 2007
Nogueira and cols., J. Nanosc. Nanotech 2009

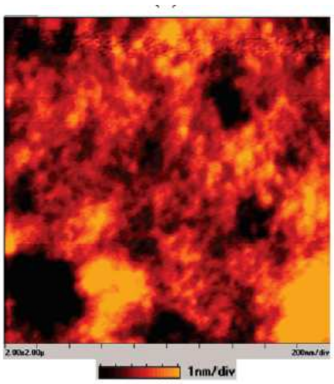


Conturbia, G. Unicamp, 2010

AFM



(b)



(c)

Before

(rms roughness = 5.3 nm)

After NT chemical modification with thiophene groups

(rms roughness = 3.6 nm)

Variation of the SWCNT content

- Addition of SWCNT increases the generation of charge carriers.
- Highest Isc and Voc values for devices containing 5 % of modified SWCNT:

Voc = 0.74 V
Isc = 9.1 μA cm^{-2}
FF = 0.29
η = 0.184 %

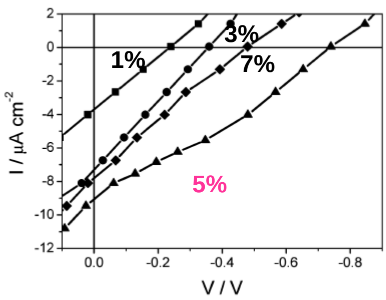
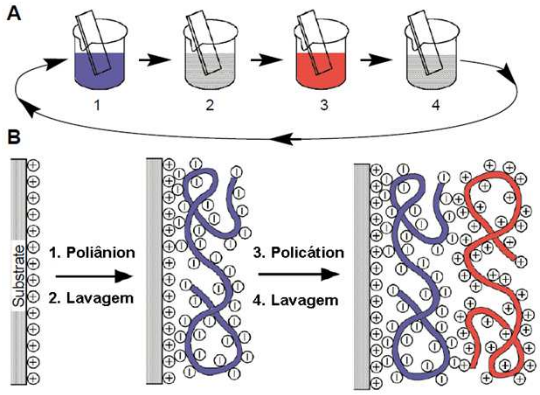


Figure 6. $I-V$ characteristics under simulated AM 1.5 illumination (Φ = 10.6 W m^{-2}) for the polymer solar cells using P3OT composites with different SWCNT-THIOP doping levels. The device configuration is the same as described in Figure 4: (■) 1 wt %; (●) 3 wt %; (▲) 5 wt %; (♦) 7 wt %.

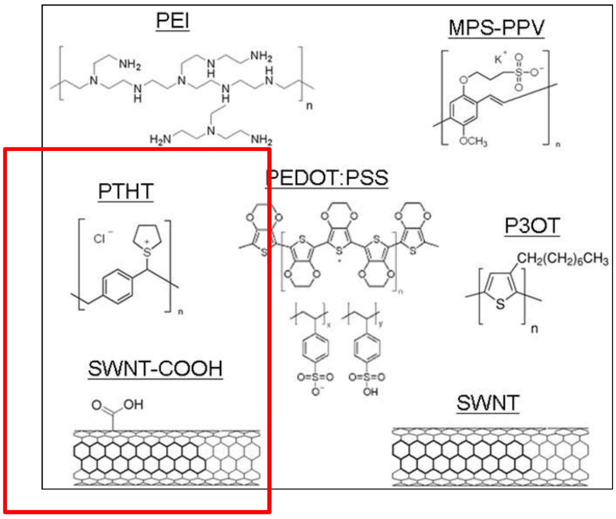
Nogueira and cols., J. Phys Chem C., 2007

Layer-by-layer technique to grow carbon based thin films for 3G solar cells

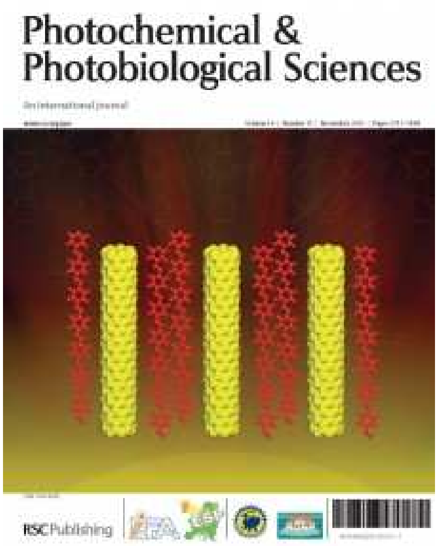


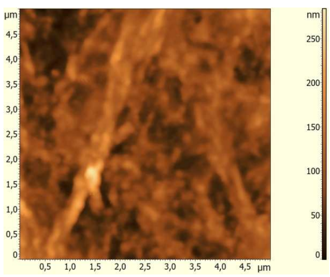
G. Decher, Science 277, 1232, 1997

- Materials used in this work



- Luiz C. P. Almeida, Doctoral Thesis

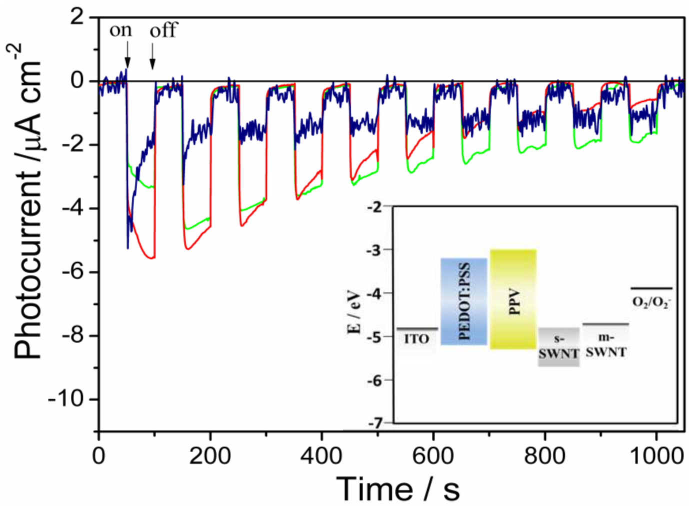


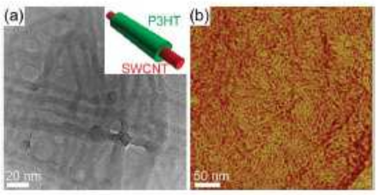


100 layers of PPV +
8 layers of SWCNT

L.C. P. Almeida et. al. J. Photochem. Photobiol., 2011

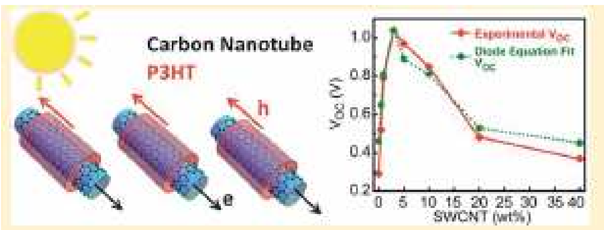
Photoelectrochemistry

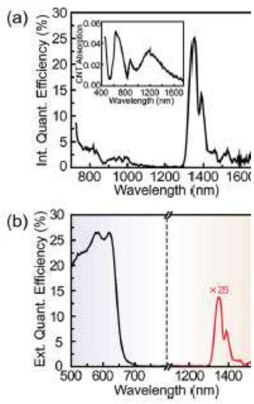




Improving…

Figure 2. (a) Bright-field TEM image of a mixture of P3HT/s-SWCNTs nanofilament sample with SWCNT concentration $x = 3$ wt %. (Inset) Schematic image of a P3HT (green)/s-SWCNT (red) nanofilament prepared in this work. (b) AFM phase image of P3HT/s-SWCNT nanofilaments in a sample with SWCNT concentration $x = 3$ wt % showing the wormlike morphology of the active layer.





dx.doi.org/10.1021/nl202796u | Nano Lett. 2011, 11, 5316–5321

Organic Photovoltaic Devices Based on a Novel Acceptor Material: Graphene**

By *Zunfeng Liu, Qian Liu, Yi Huang, Yanfeng Ma, Shougen Yin,** *Xiaoyan Zhang, Wei Sun,* and *Yongsheng Chen**

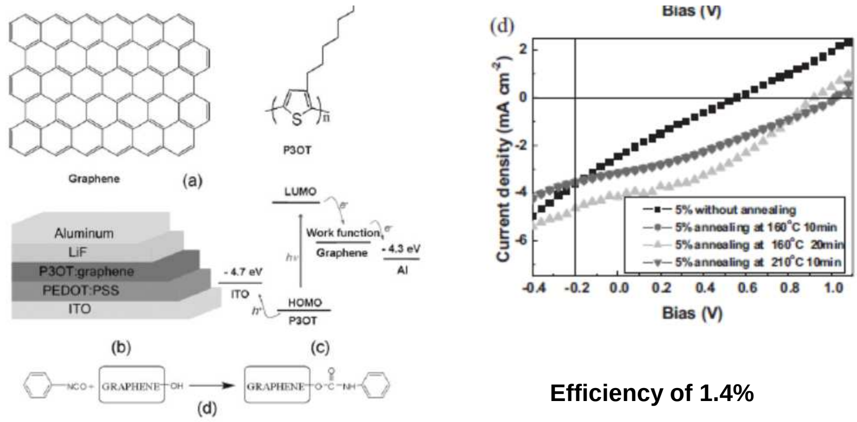


Figure 2. a) The idealized chemical structures of graphene and P3OT. b) Schematic of the device with P3OT/graphene thin film as the active layer and the structure ITO (ca. 17 Ω sq^{-1})/PEDOT:PSS (40 nm)/P3OT:graphene (100 nm)/LiF (1 nm)/Al (70 nm). c) Energy level diagram of P3OT and SPFGraphene. d) Schematic representation of the reaction of phenyl isocyanate with graphene oxide to form SPFGraphene.

Efficiency of 1.4%

Limitations in NanoPV

- Low absorption beyond 600 nm (we need novel dyes, conducting polymers, etc with better absorption)
- Low charge carrier mobility for organic semiconductors. Morphology control is also a problem in these devices.
- Liquid electrolyte in DSSC causes leakage and compromises stability.
- Need of novel free-iodine redox electrolytes (DSSC)
- More stability tests are required for both technologies

Perspectives

- Nanomaterials are strong candidates in solar energy conversion, specially for DSC and OPV cells;
- Both cells have their future guaranteed in the PV market; specially because their versatility and cost;
- For DSC, novel solid electrolytes, novel redox systems can be an alternative;
- Chemists can find their place in preparing more efficient dyes or polymers that absorb beyond 600 nm
- The tendency is towards more organized nanostructured devices, use of nanotubes, nanowires to improve the electron transport

LNES team

❋Dye sensitized Solar Cells
❋ Organic and Hybrid Solar Cells
❋ SC photocatalysis for CO_2 reduction and H_2 production

Acknowledgements

Nanoenergy Cyted network promotes cooperation among countries in Latin America (coordinated by Juan Bisquert)

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Obrigado!