

Mesoscopic Injection Solar Cells for Electricity Generation from Sunlight

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**Michael Graetzel
Swiss Federal Institute of Technology Lausanne
michael.graetzel@epfl.ch**

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Industrial Partners,



Humanity's Top Ten Problems for next 50 years

1. ENERGY
2. WATER
3. FOOD
4. ENVIRONMENT
5. POVERTY
6. TERRORISM & WAR
7. DISEASE
8. EDUCATION
9. DEMOCRACY
10. POPULATION



2003	6.3	Billion People
2050	8-10	Billion People

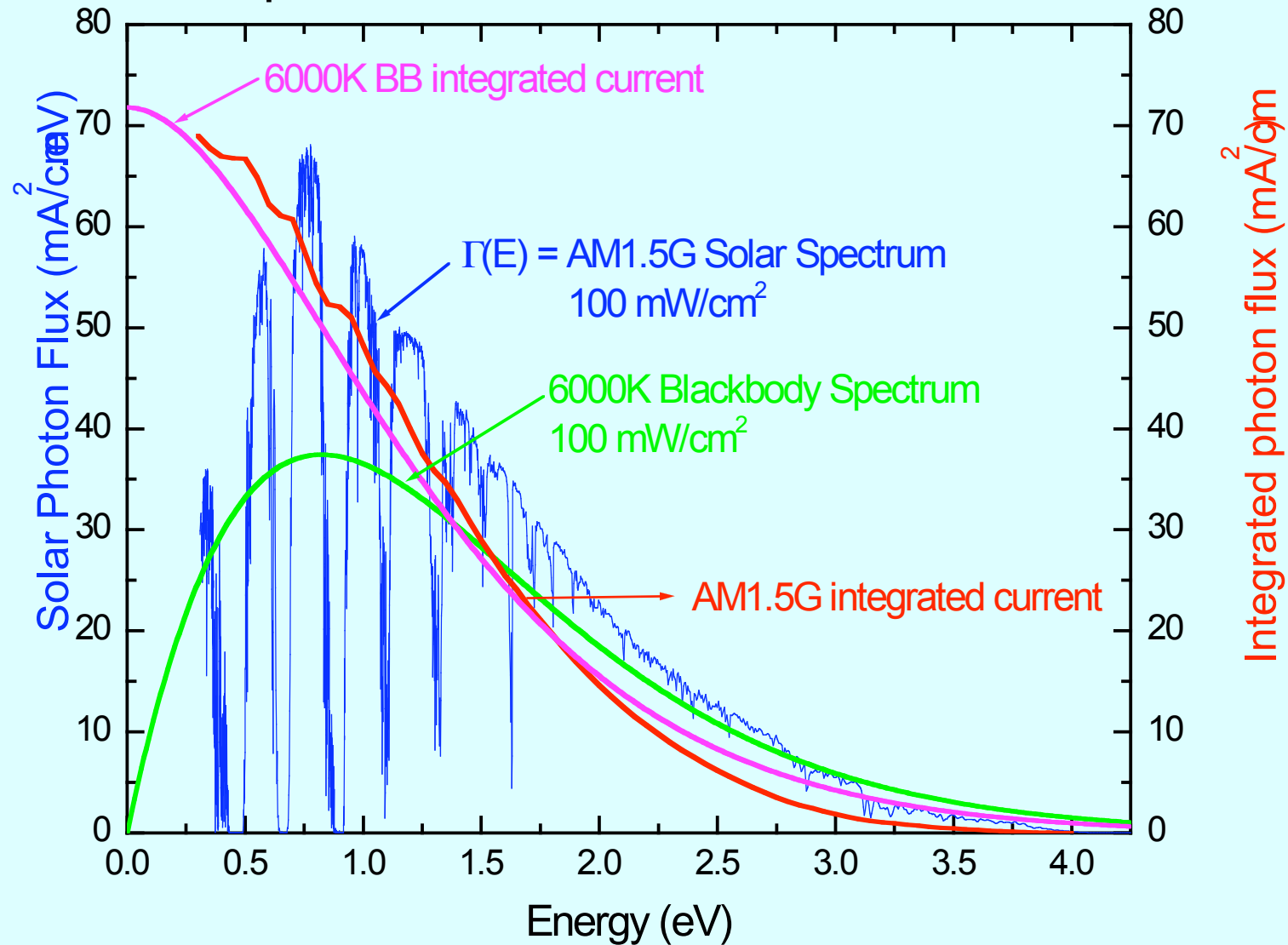
Source Richard Smalley Energy & Nanotechnology Conference
Rice University, Houston May 3, 2003

THE SOLAR CHALLENGE

- **With a projected global population of 12 billion by 2050 coupled with moderate economic growth, the total global energy consumption is estimated to be ~28 TW. Current global use is ~11 TW.**
- **To cap CO₂ at 550 ppm (twice the pre-industrial level), most of this additional energy needs to come from carbon-free sources.**
- **Solar energy is the largest non-carbon-based energy source (100,000 TW).**
- **However, it has to be converted at reasonably low cost.**

THE SOLAR RESOURCE

Solar Spectrum and Available Photocurrent



Photovoltaic Cells

I. 1st Generation

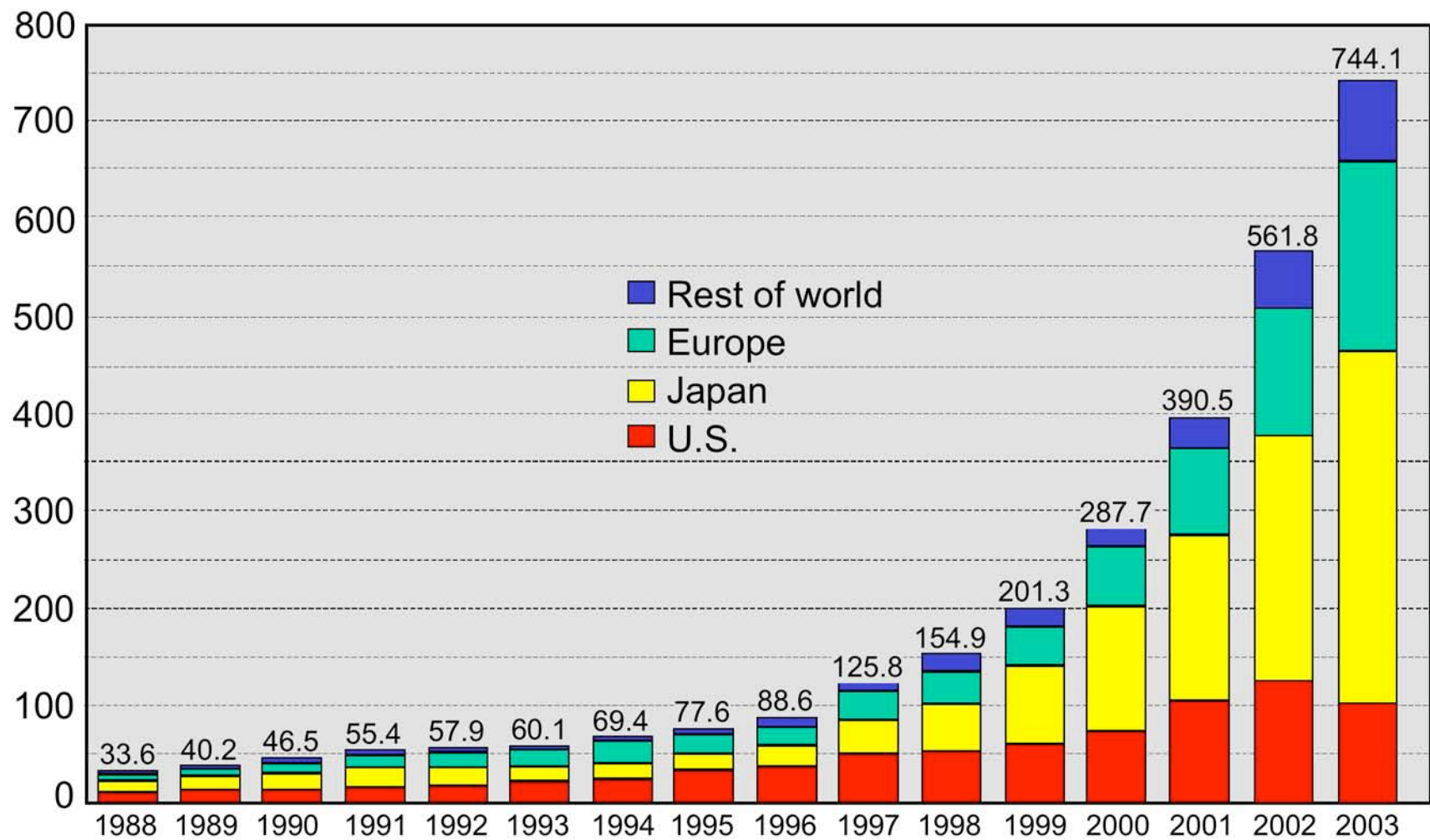
- Single crystal Si
- Poly-grain Si

II. 2nd Generation (Low Cost--Mainly Thin Films)

- Amorphous Si
- Thin film Si
- CuInSe_2
- CdTe
- Dye-sensitized Photochemical Cell
- Organic PV (molecular and polymeric)

III. 3rd Generation ($n_{\text{theor}} > 31\%$ (the Queisser-Shockley limit))

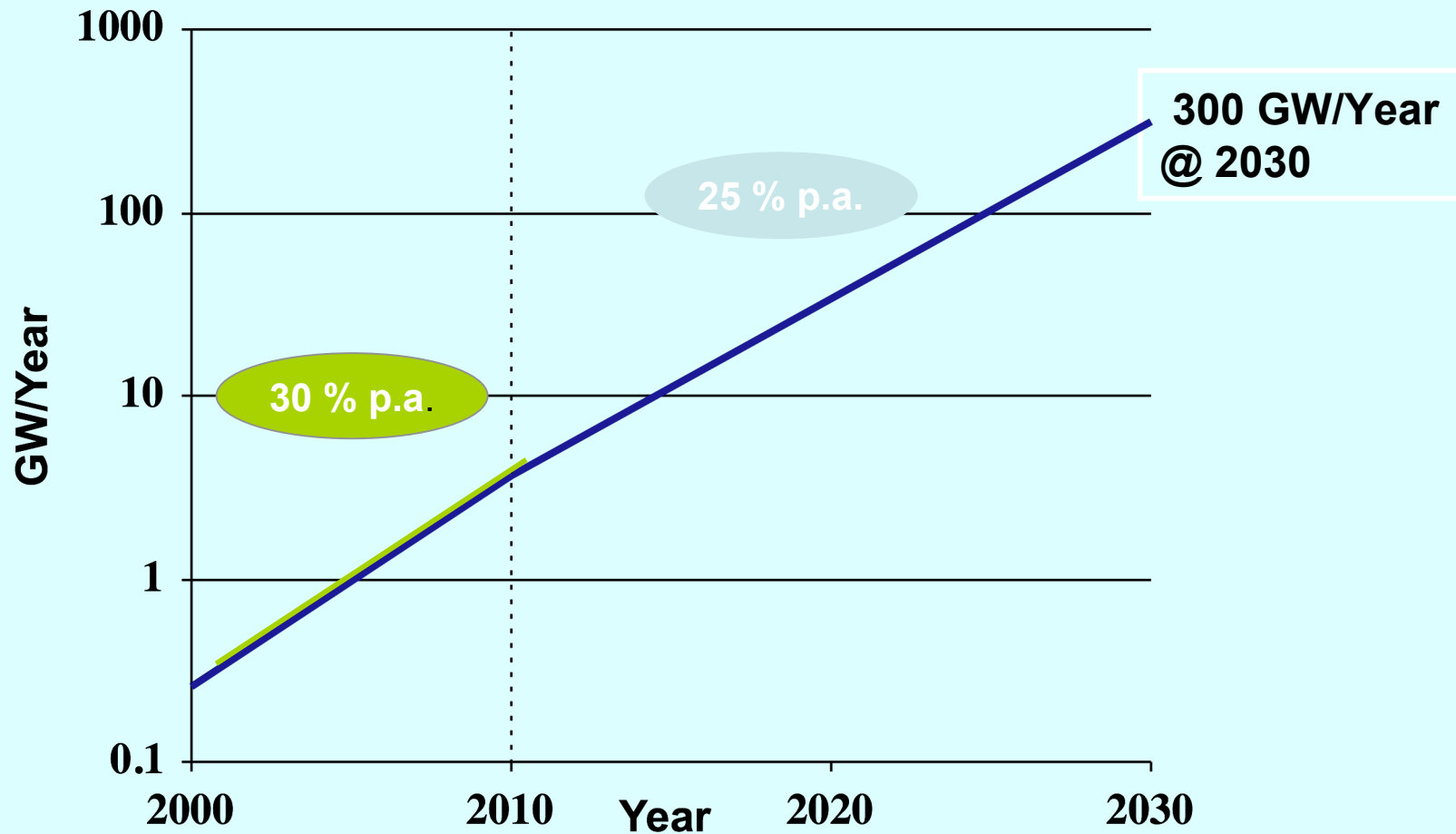
- *(High efficiency multi-gap tandem cells (already here))*
- Hot electron converters
- Carrier Multiplication cells
- Mid-band PV
- Quantum Dot Solar Cells
- Other approaches



Source: *PV News*, March 2004

World PV Cell/Module Production (in MW)

Photovoltaic market growth projection until 2030



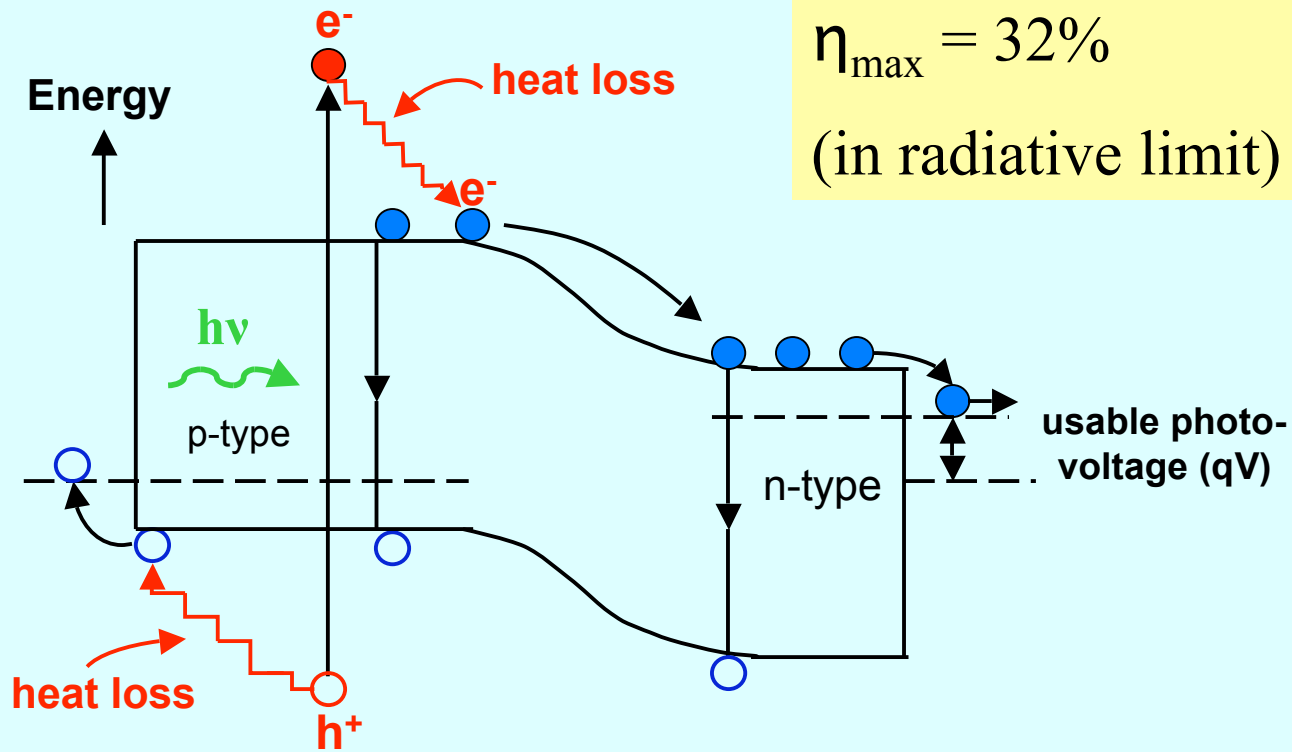
Courtesy Dr. Winfried Hoffman, CEO, RWE, SCHOTT Solar GmbH

PRESENT PV TECHNOLOGY

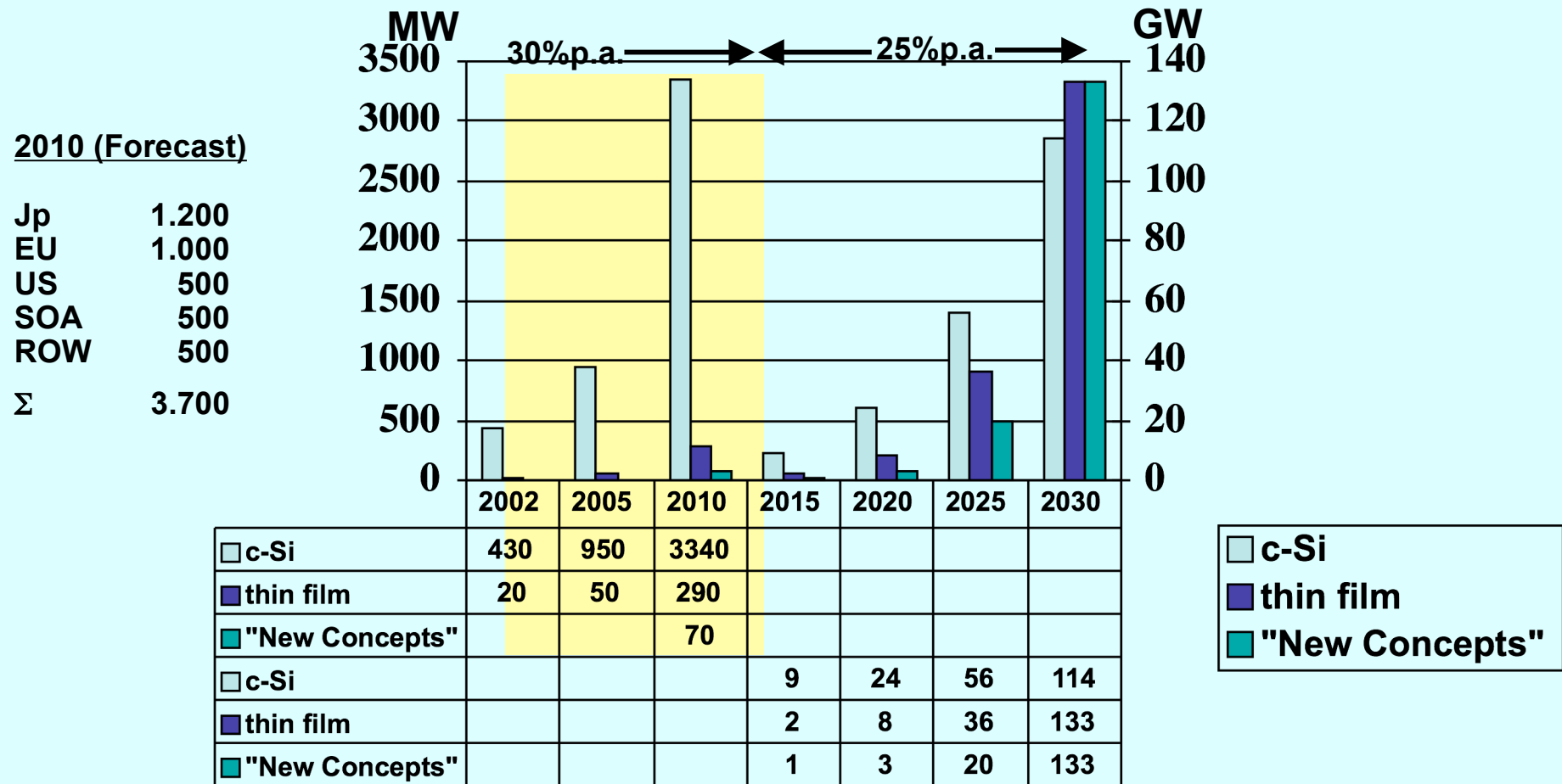
(Dominated by semiconductor p-n
junctions)

Conventional Single Homojunction PV Cell

1 e⁻ - h⁺ pair/photon; full hot carrier relaxation



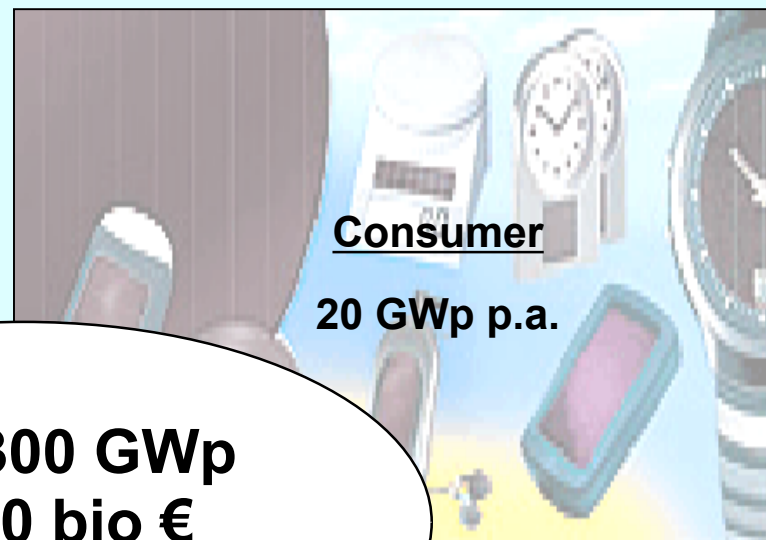
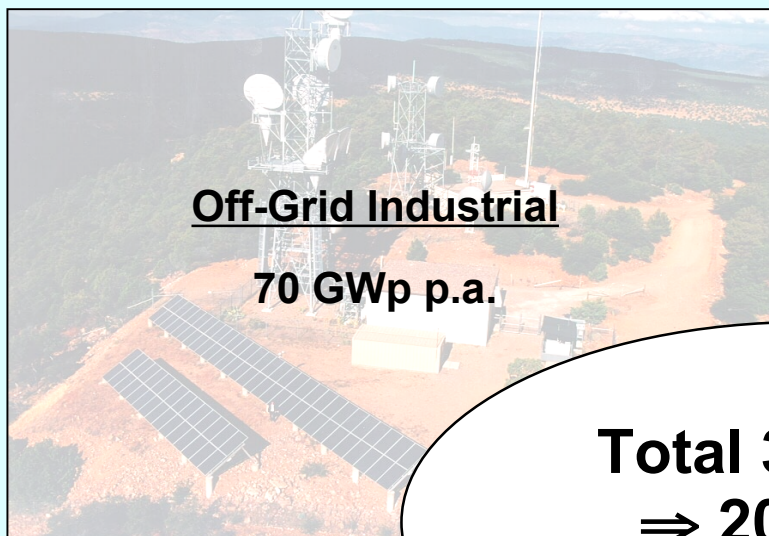
Production Forecast of Solar Modules Using Different Technologies



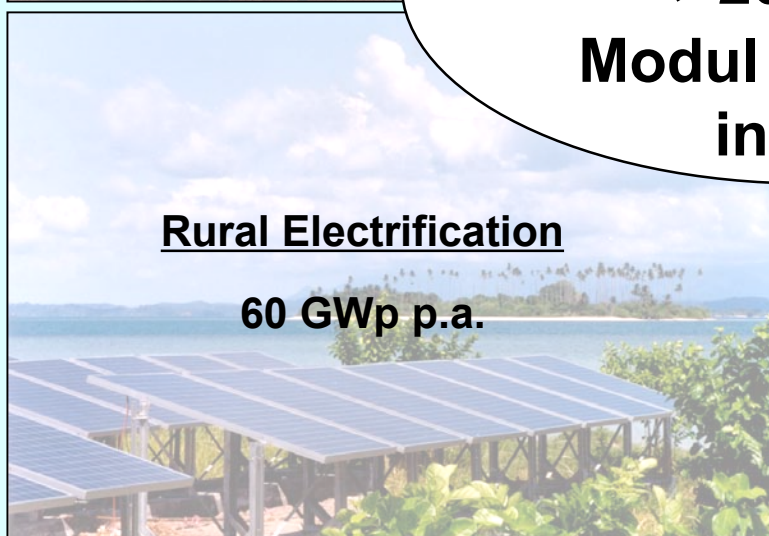
Courtesy Dr. Winfried Hoffman, CEO, RWE, SCHOTT Solar GmbH

Market Size in 2030 for the four market segments

Courtesy Dr. Winfried Hoffman, CEO, RWE, SCHOTT Solar GmbH



**Total 300 GWp
⇒ 200 bio €
Modul Turnover
in 2030**



Emerging and new applications call for:

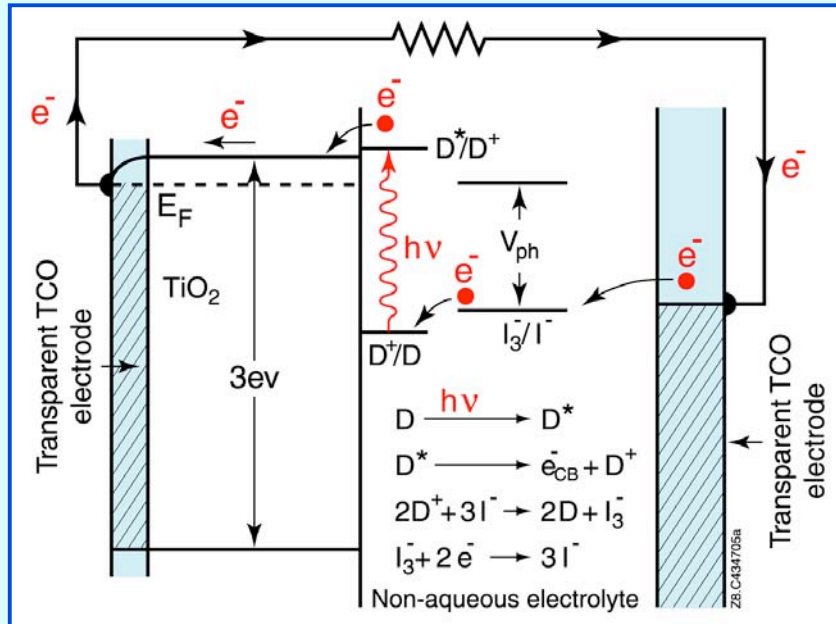
- colour
- flexibility
- light weight
- easy of integration
- many more

... further development and new technologies in order to meet optimally the customer demands and needs

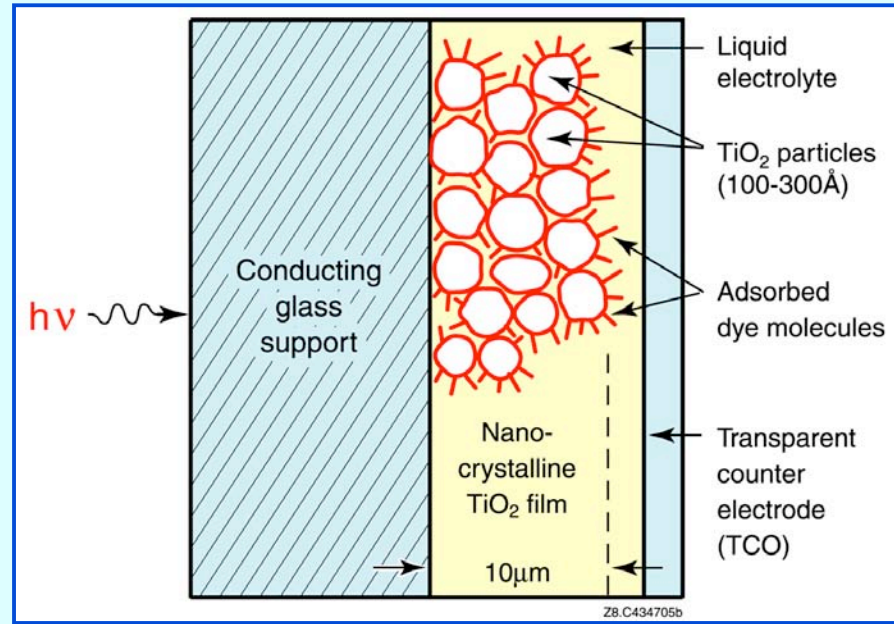
Courtesy Dr. Winfried Hoffman, CEO, RWE, SCHOTT Solar GmbH

Mesoscopic Injection Solar Cells

Band Diagram

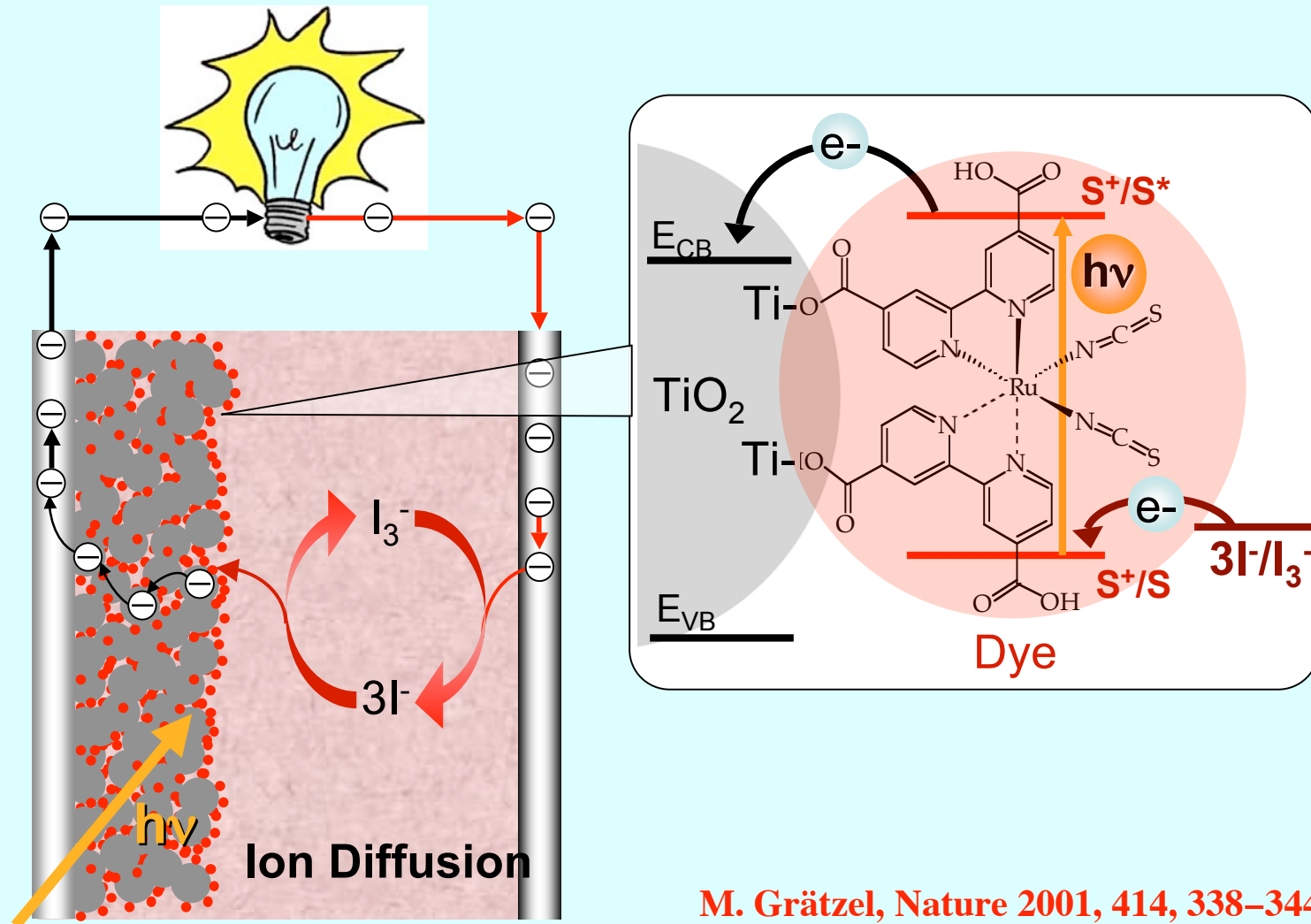


Scheme



M. Grätzel, Nature 2001, 414, 338-344

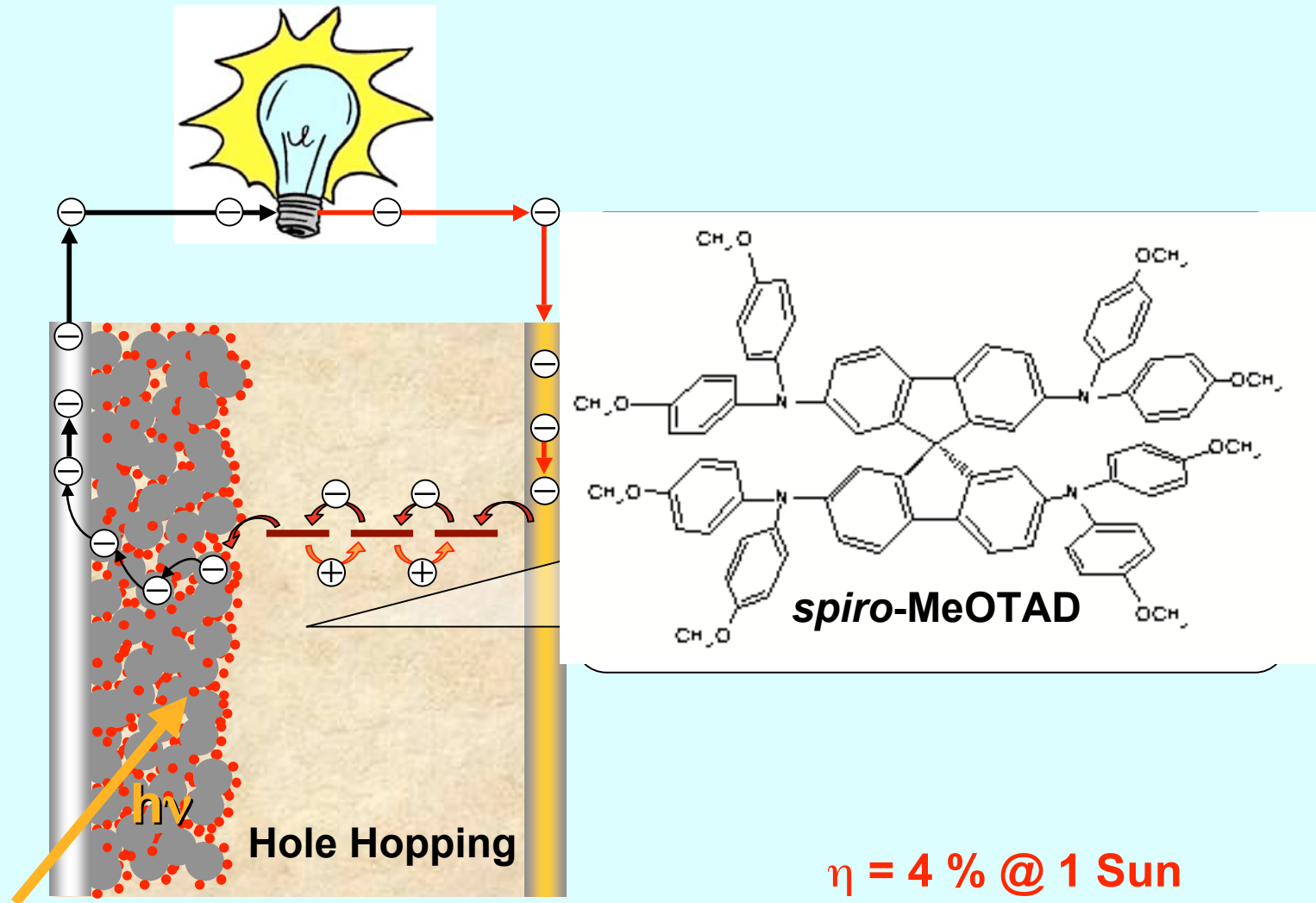
Sensitized mesoscopic heterojunctions



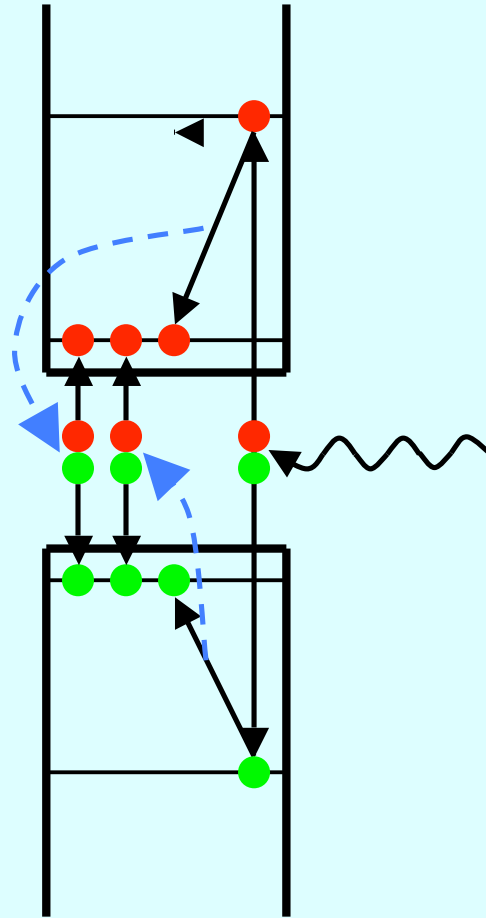
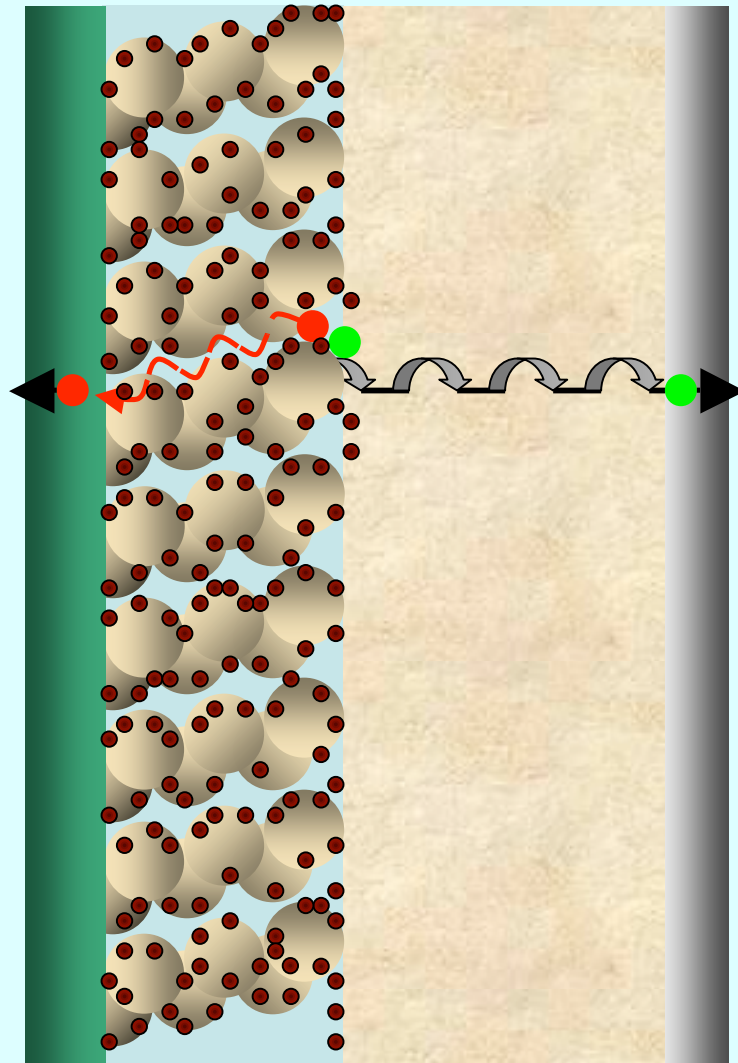
M. Grätzel, Nature 2001, 414, 338–344.

B. O'Regan, M. Grätzel, Nature 1991, 353, 737–740

Device Concept: "solid-state"

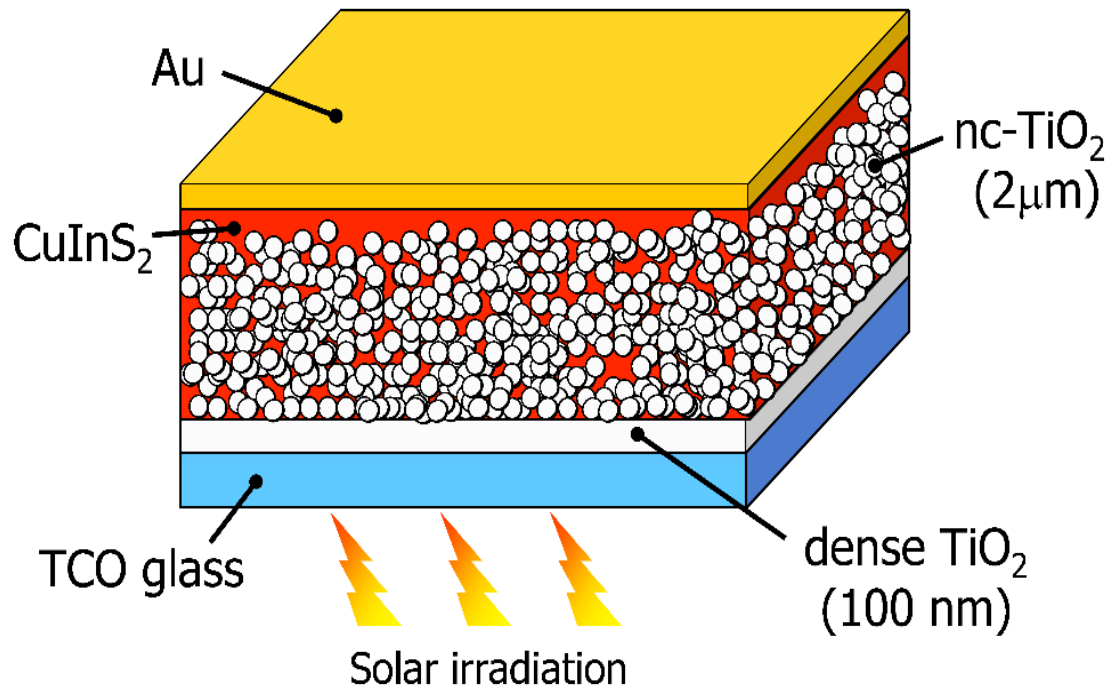


U. Bach, D. Lupo, P. Comte, J.-E. Moser, F. Weissörtel, J. Salbeck, H. Spreitzer and M. Grätzel, NATURE 395, 583-585 (1998)



**R.D. Schaller and V.I. Klimov, Phys. Rev. Letts, 92,
186601 (May), 2004 (PbSe QDs)**

3D solar cells

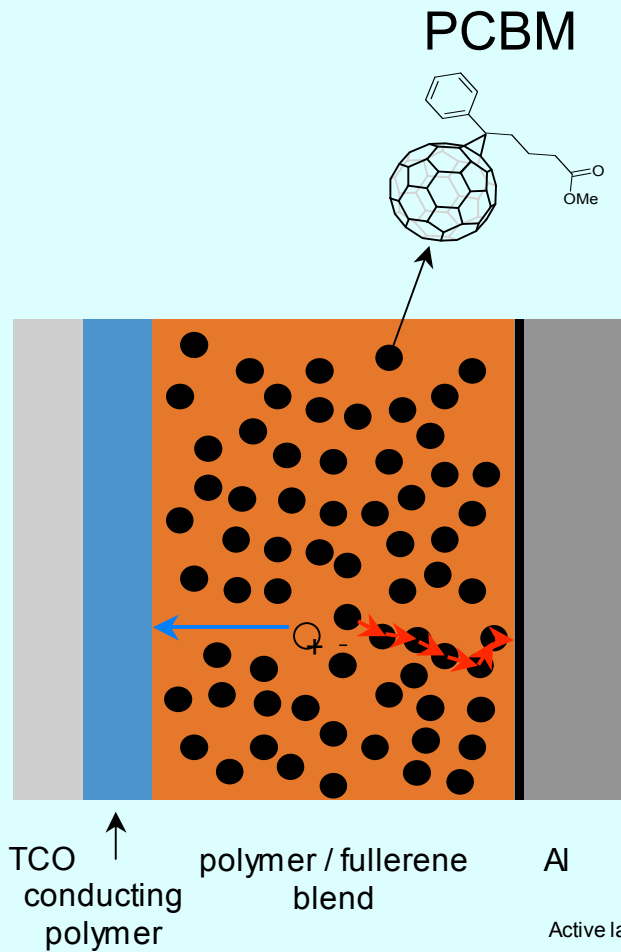


- 1 percolation of electrons and holes
- 2 selective contacts
- 3 no electrical shunts (pinholes)
- 4 stoichiometric TiO_2
- 5 p-type CuInS_2
- 6 buffer layer
- 7 intimate contact
- 8 complete filling of pores

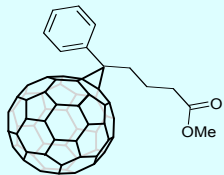
M. Nanu, J. Schoonman, and A. Goossens, Advanced Materials 16 (2004) 453

M. Nanu, J. Schoonman, and A. Goossens, Adv. Func. Mat. 15 (2005) 95

Bulk organic heterojunction solar cell

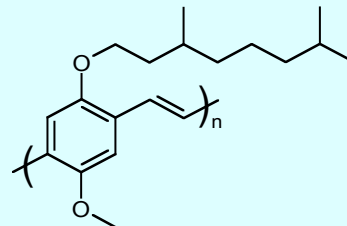


PCBM

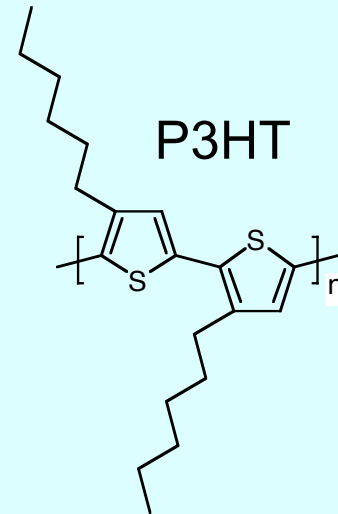


blended with:

OC₁C₁₀-PPV



P3HT



$$J_{SC} = 5.3 \text{ mA/cm}^2$$

$$V_{OC} = 0.82 \text{ V}$$

$$FF = 61\%$$

$$\eta = 2.5\%$$

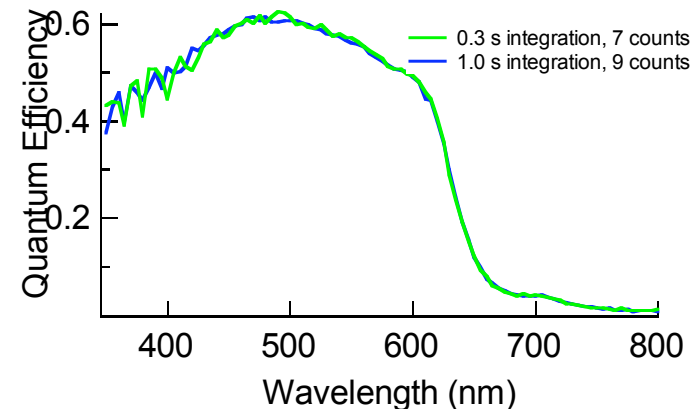
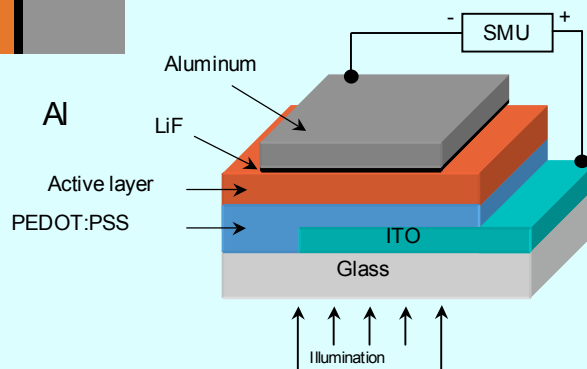
$$J_{SC} = 9.3 \text{ mA/cm}^2$$

$$V_{OC} = 0.56 \text{ V}$$

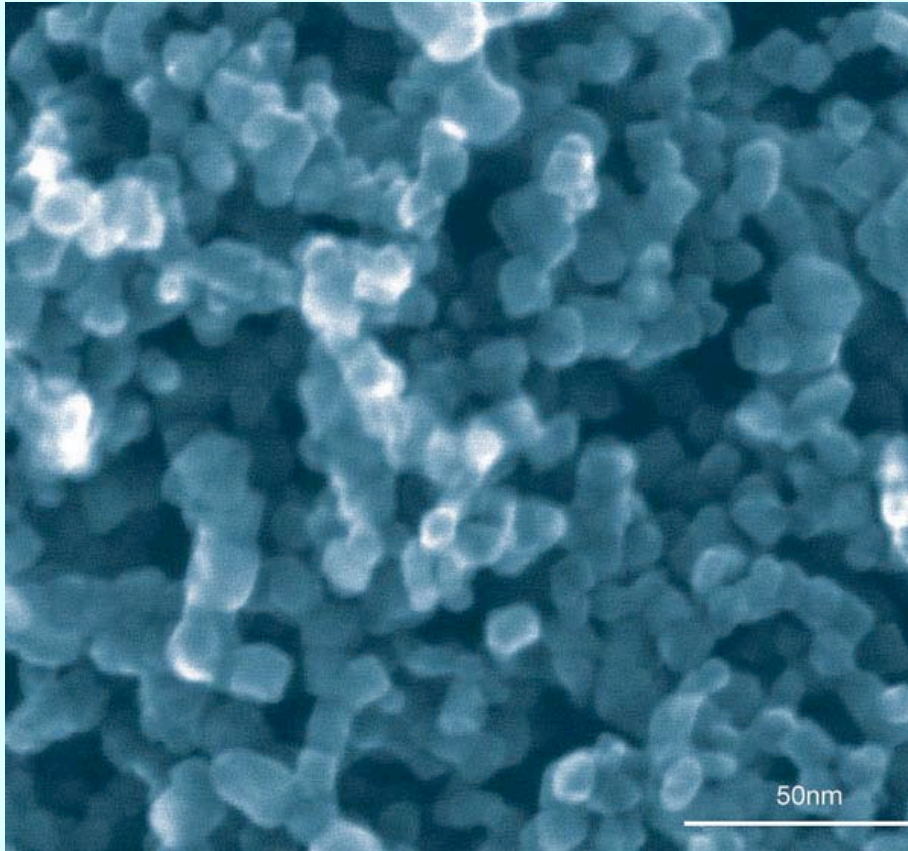
$$FF = 60\%$$

$$\eta = 3.5\%$$

(under AM1.5 illumination)



From G. Rumbles NREL



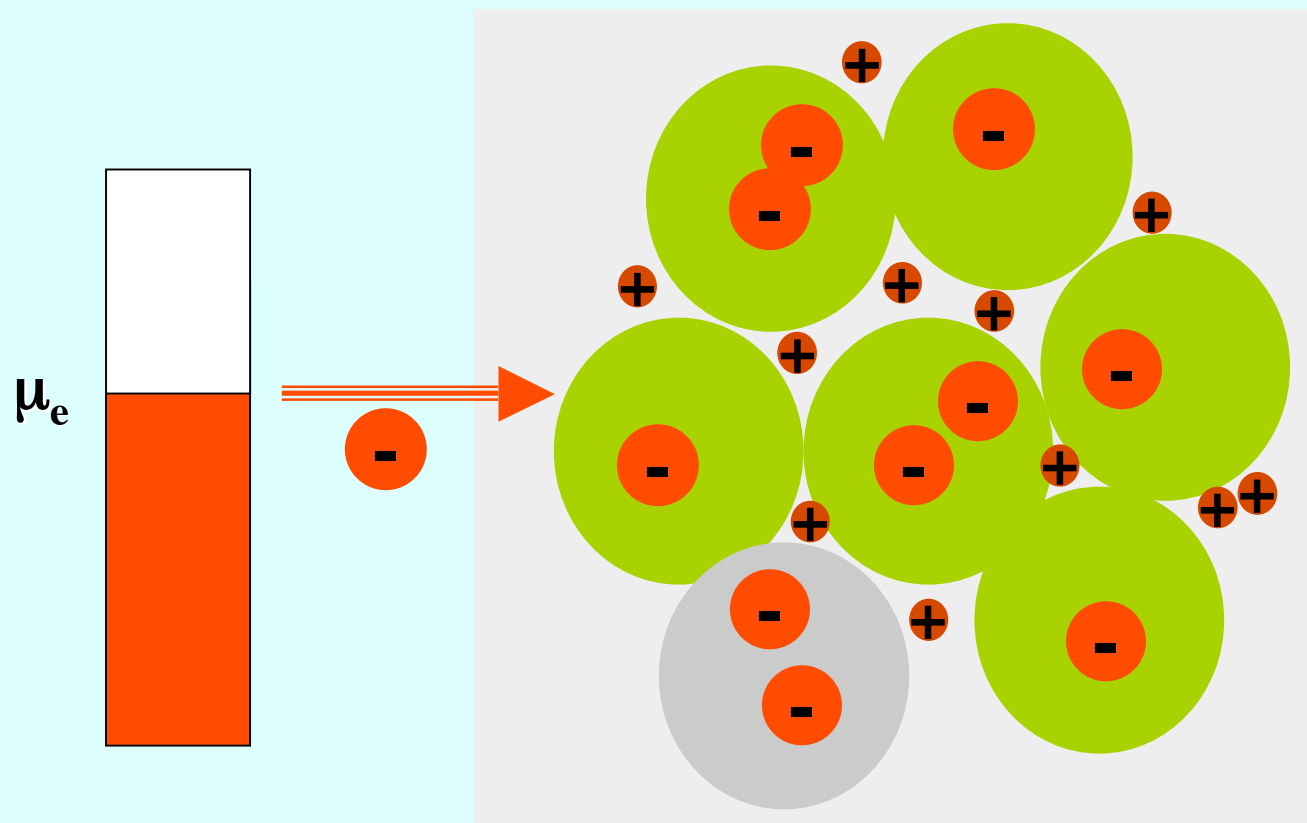
Outline

- **Mesoporous junctions, interfacial and cross-surface charge transfer**
- **Photoinduced charge separation**
- **Photogalvanic generation of electricity from sunlight**

Mesoscopic semiconductor films exhibit extraordinary properties

- surface amplification ca 100 times for each micron film thickness
- **Interpenetrating network electronic junction having huge contact area**
- ease of electron percolation through the particle network
- very rapid lithium insertion and release
- high photocatalytic activity
- high sensitivity for detecting ambients
- efficient photovoltaic energy conversion

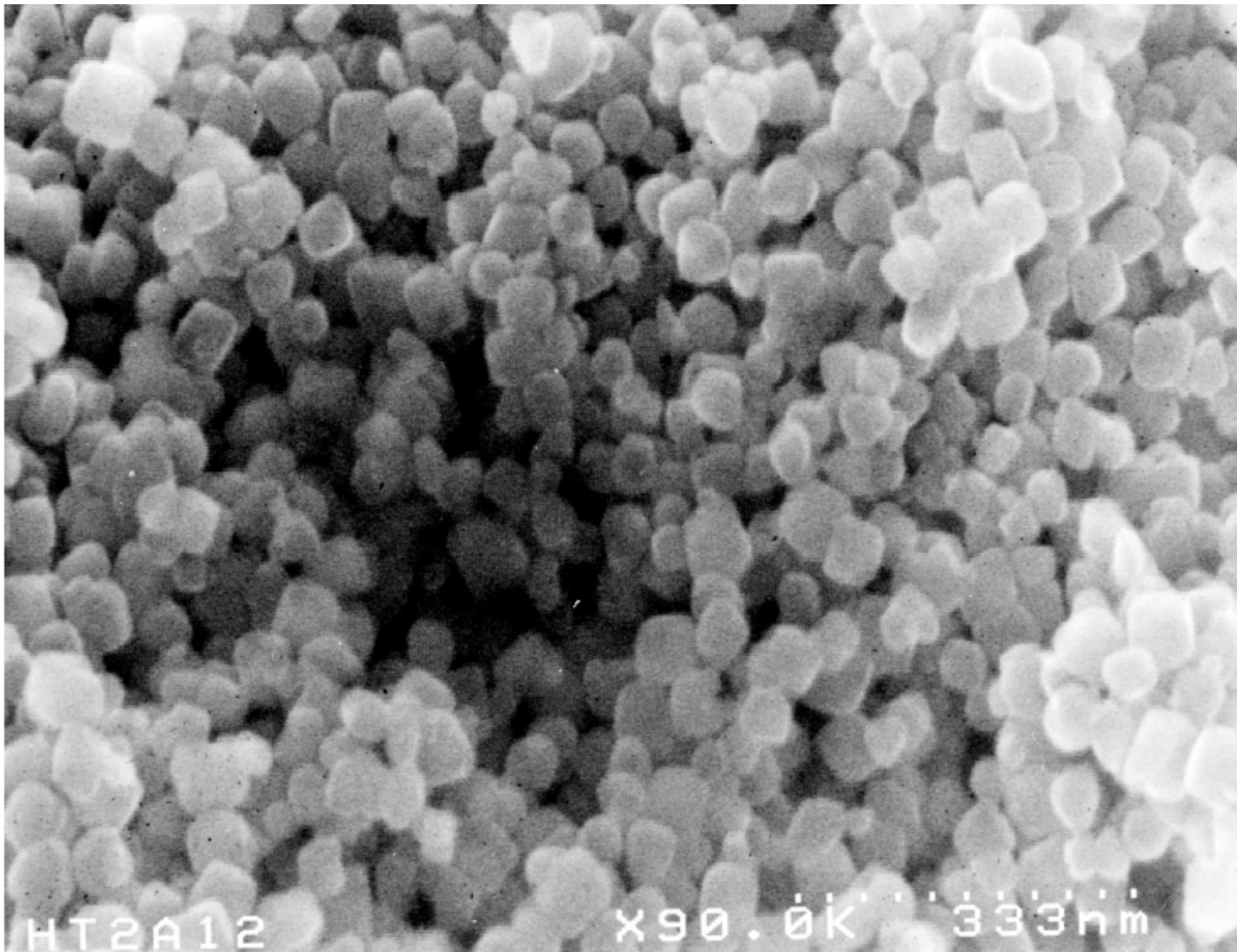
Rapid electron percolation through nanocrystals



Charge of electrons compensated by inert positive ions in electrolyte

No space charge limitation of current !

G. Rothenberger, M Grätzel and D Fitzmaurice, *J.Phys Chem.* **1992**,96,5983



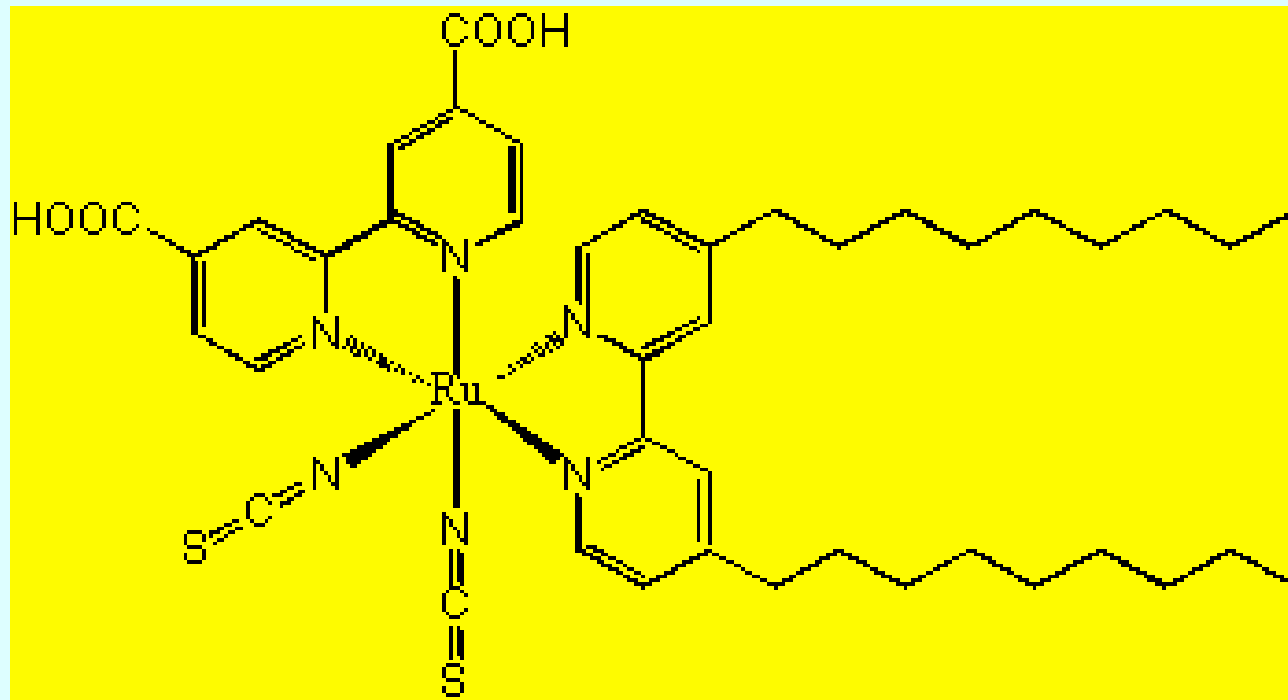
Kavan M. Grätzel *Electrochemical and Solid State Letters* 5 (2): A39-42 (2002)

Molecular wiring of insulating Nanocrystals

**Cross surface electron and hole
transfer in self-assembled molecular
charge transport layers**

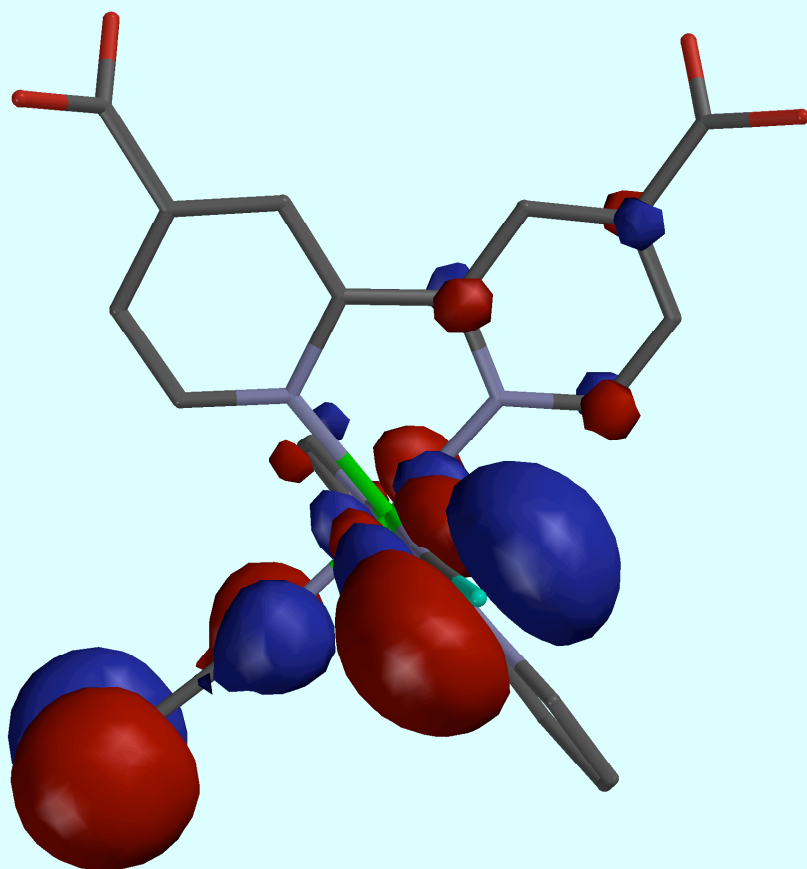
**Ambipolar lateral charge percolation
in self-assembled monolayers on
nanocrystalline insulator films**

Z907 sensitizer

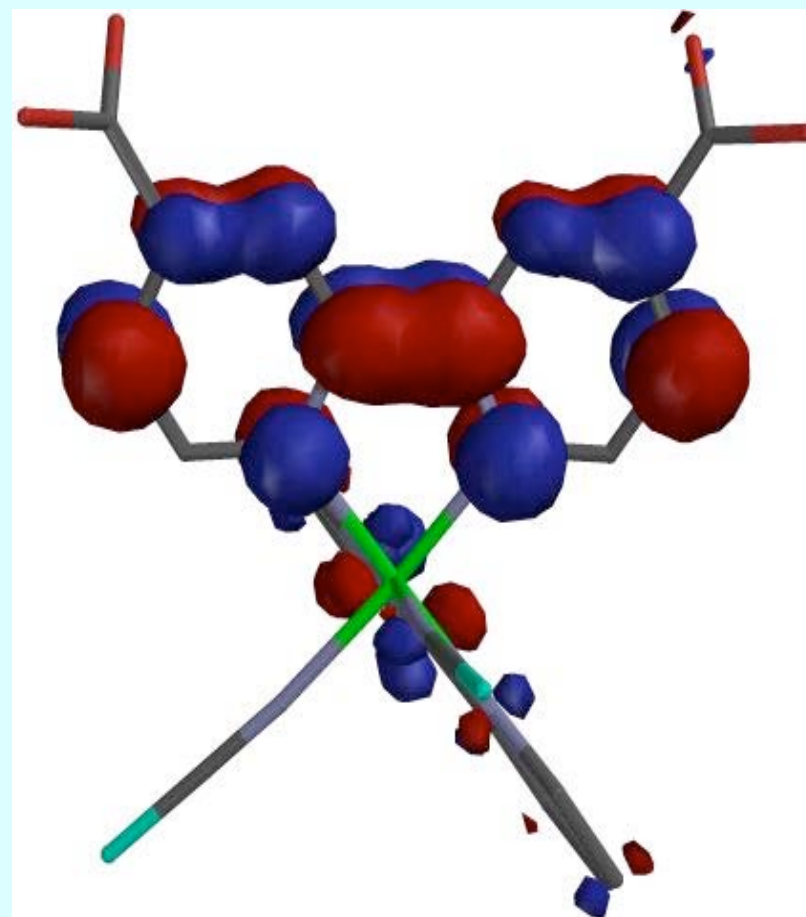


P.Wang, S.M. Zakeeruddin, R. Humphry-Baker, J.-E. Moser, M. Grätzel
Adv. Materials, 15, No. 24, 2101-2104 (2003)

HOMO

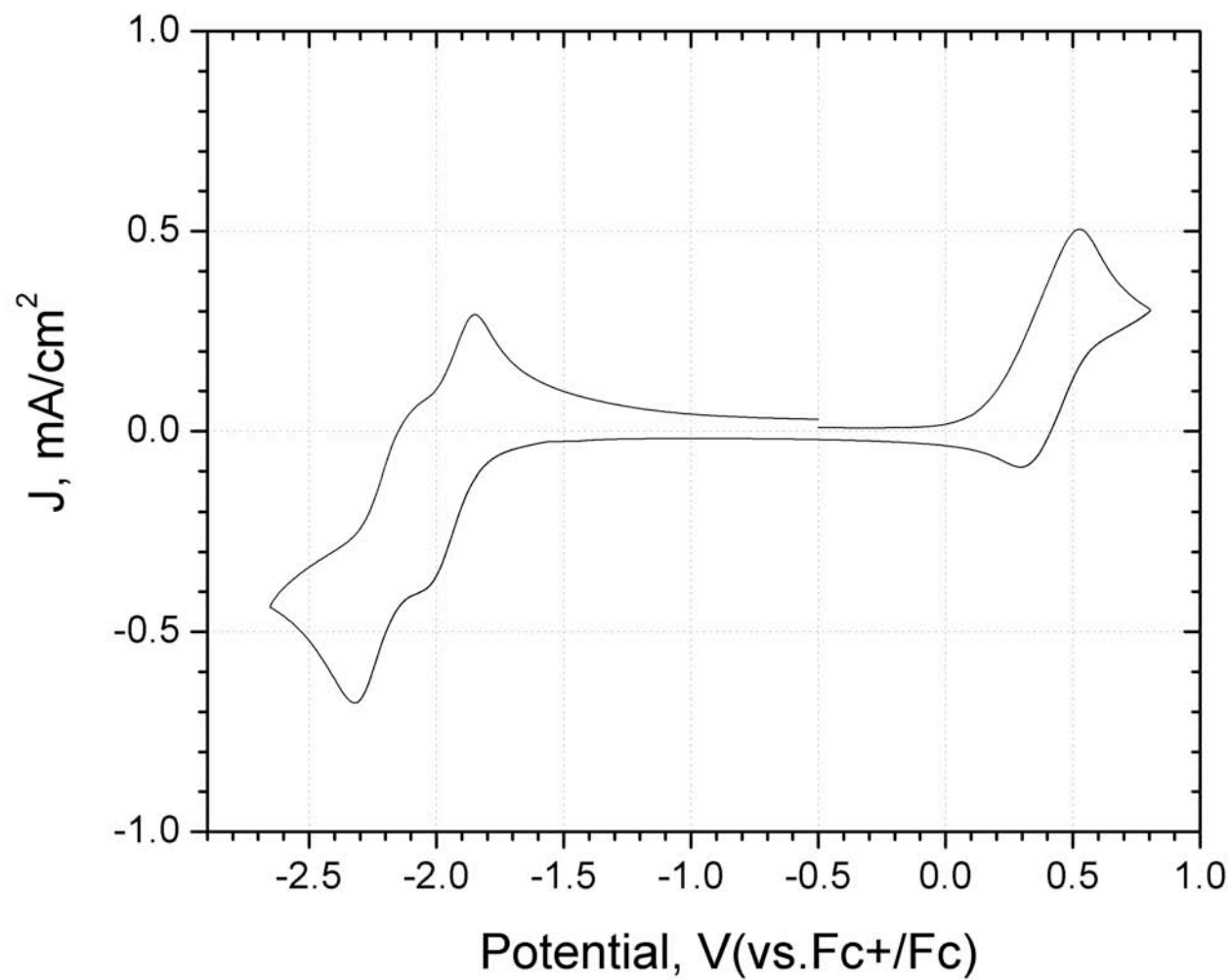


LUMO



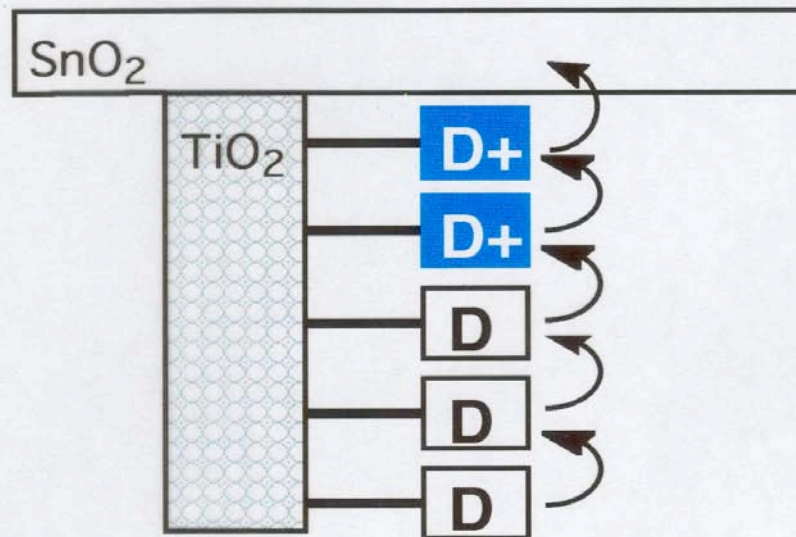
A. Hagfeldt, M. Grätzel, *Acc. Chem. Res.* 2000, 33, 2679–27

Cyclic Voltammogram of Z 907 on Aluminium Oxide Film



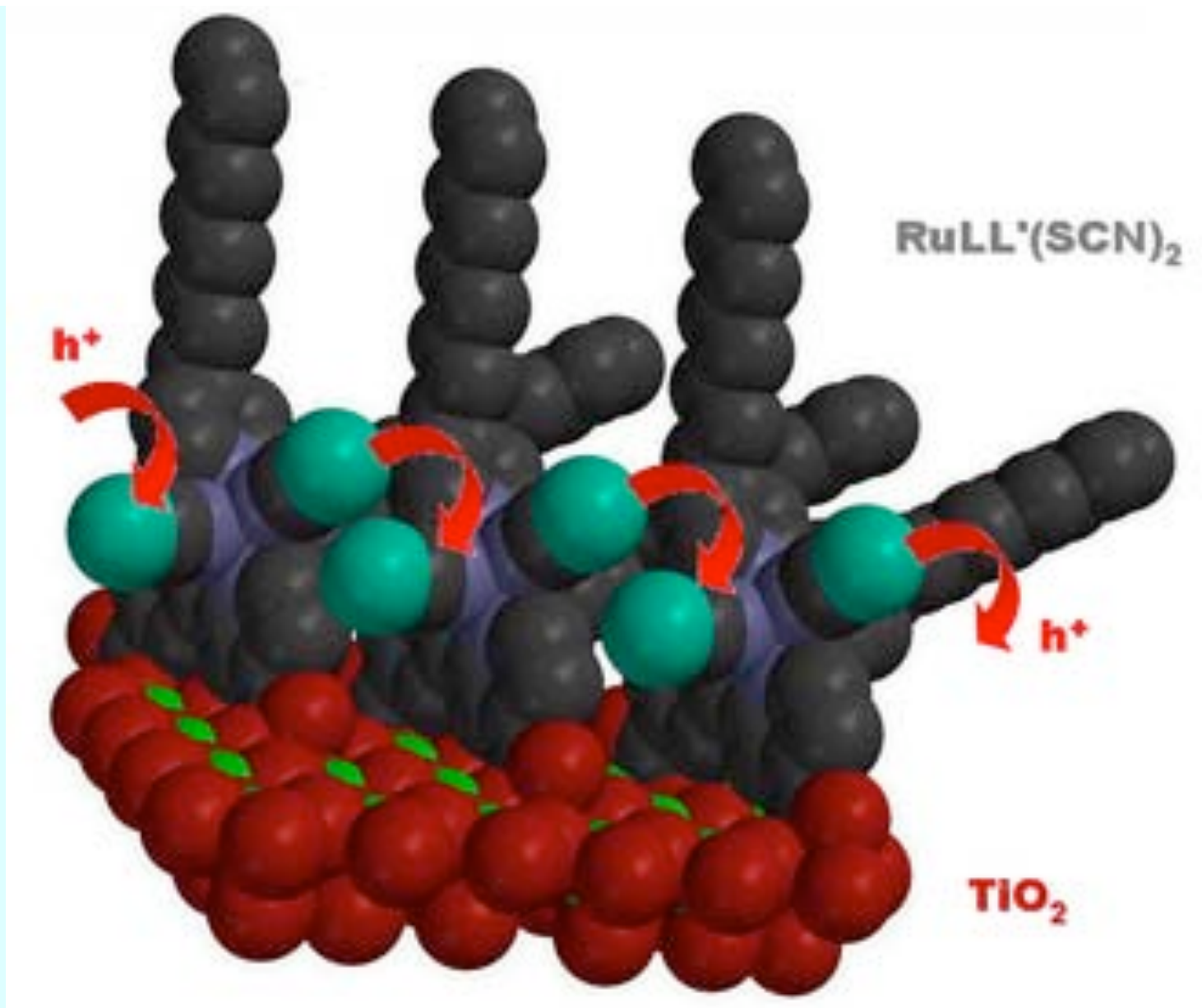
Electrolyte; EMITFSI in Acetonitrile, Scan rate= 0.1 V Sec⁻¹

2. Hole injection from the conductive support followed by lateral charge transport inside the monolayer

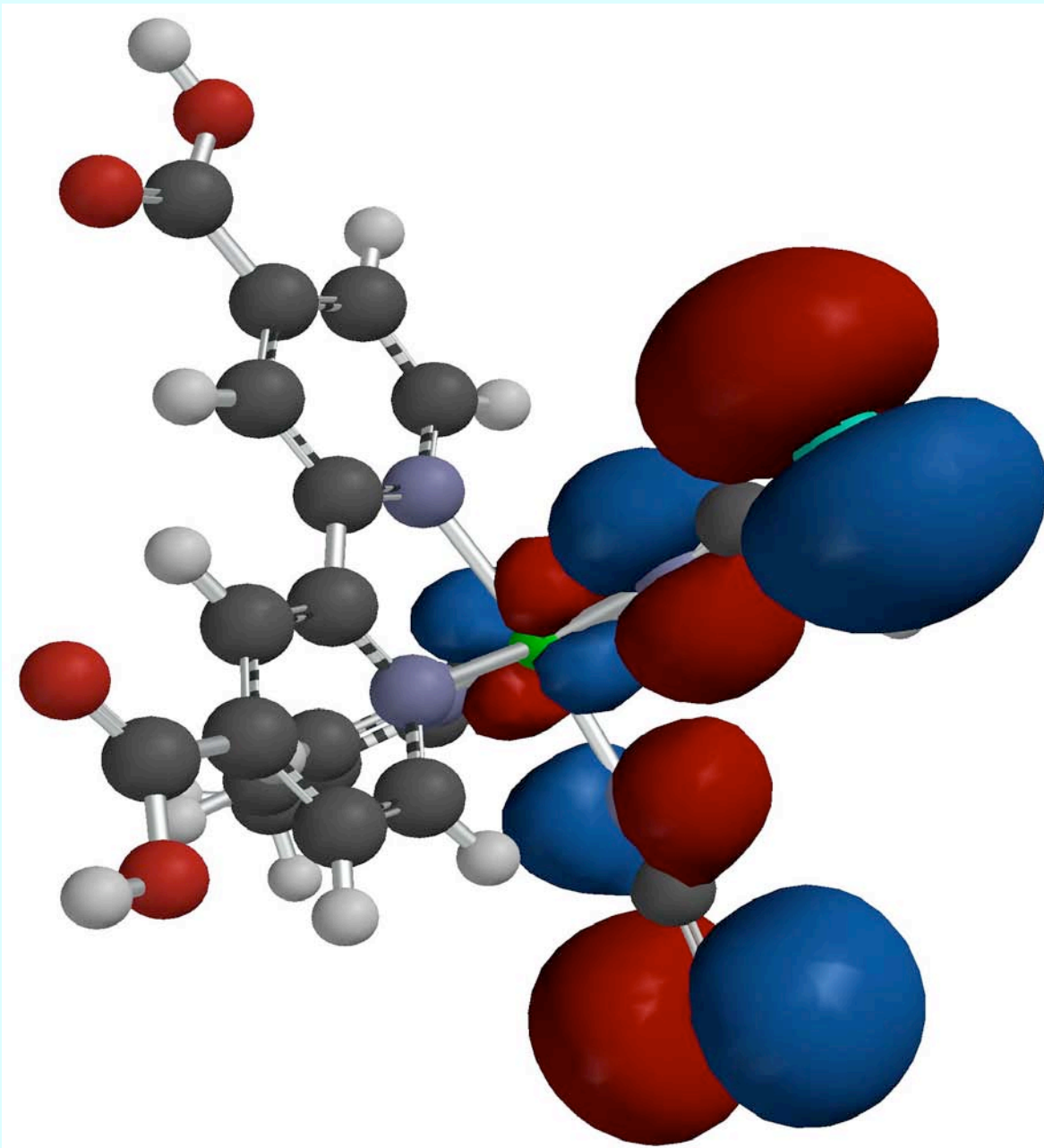


Properties:

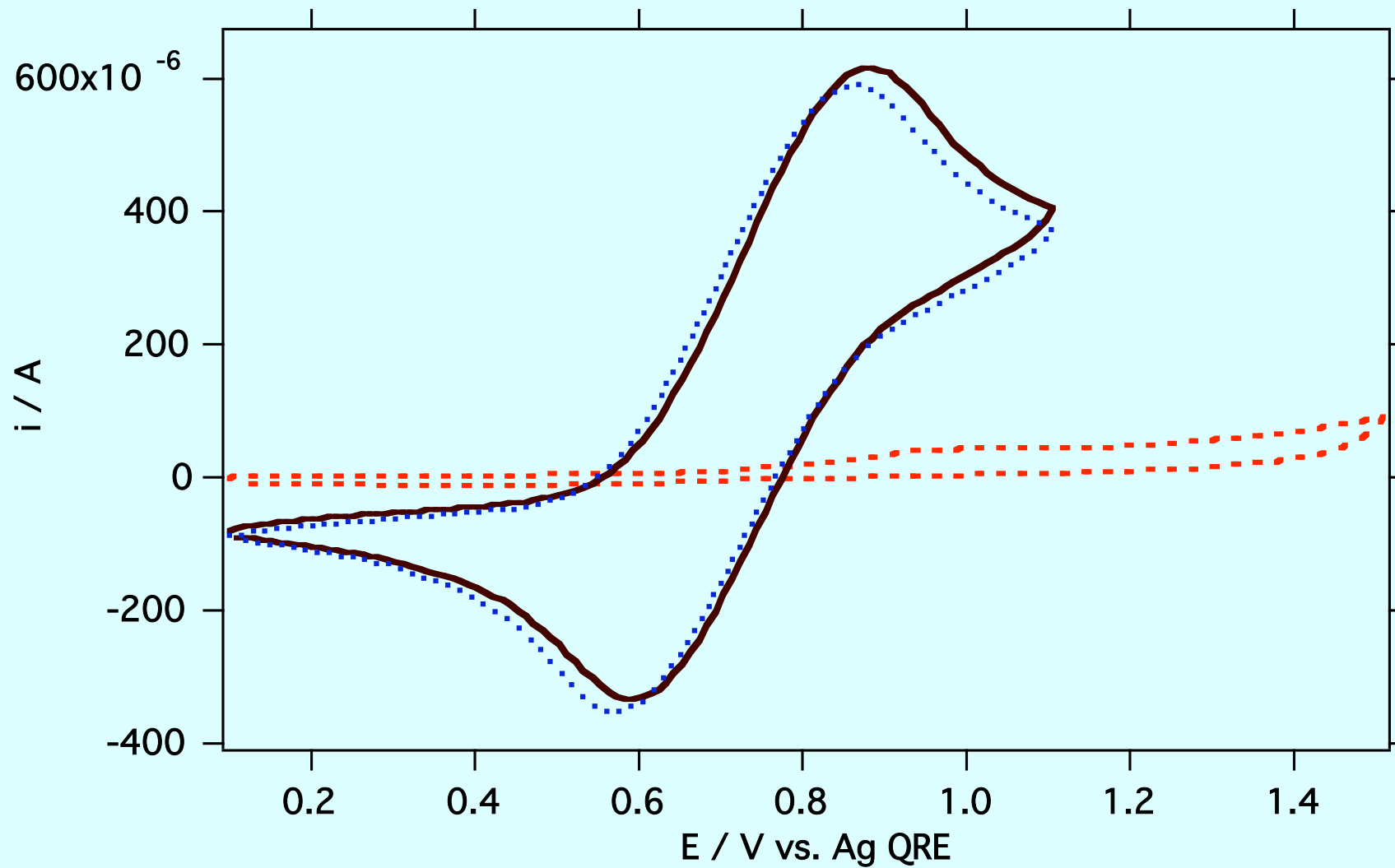
- D_{app} dependent of the surface concentration
- Percolation thershold



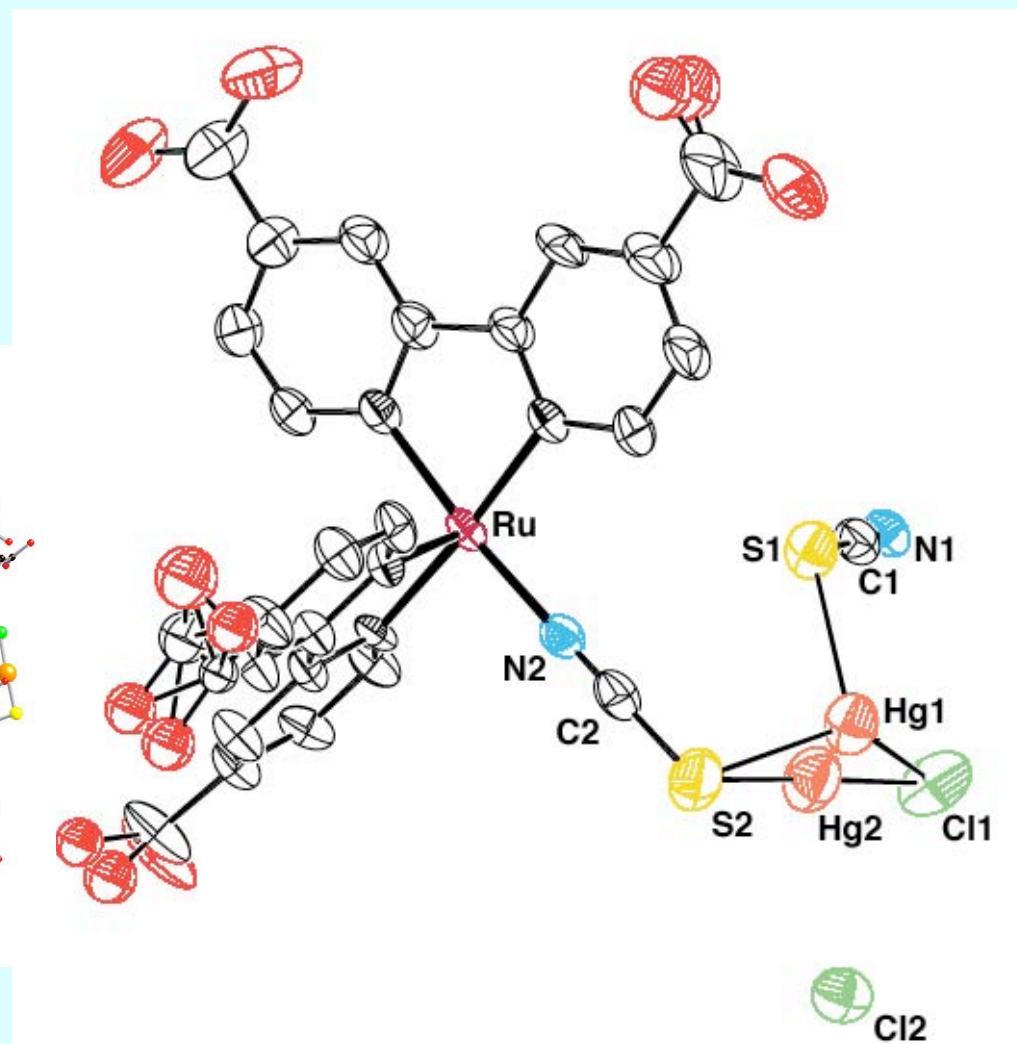
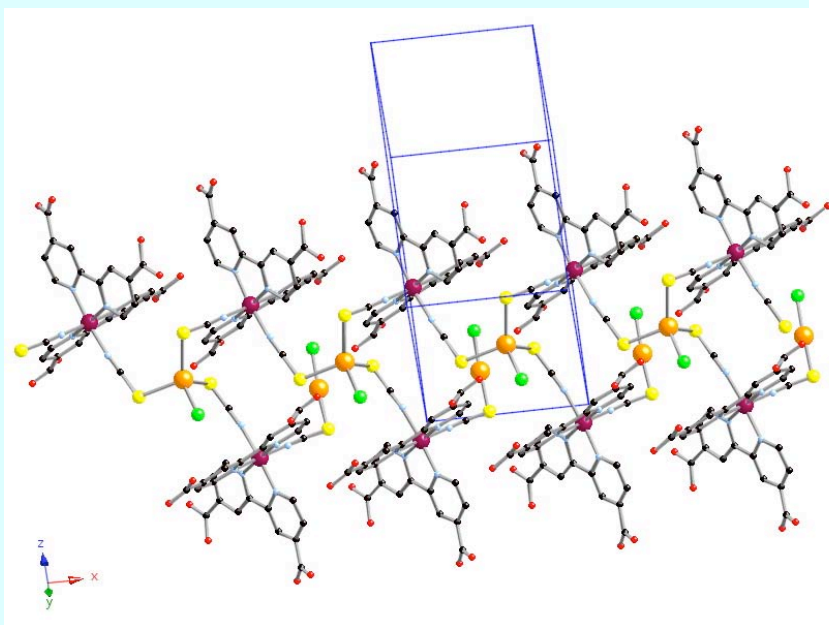
Cross surface hole percolation through a self-assembled Z-907 monolayer adsorbed on mesoscopic anatase TiO_2 .
QING WANG, ROBIN HUMPHRY BAKER AND MICHAEL GRAETZEL to be submitted

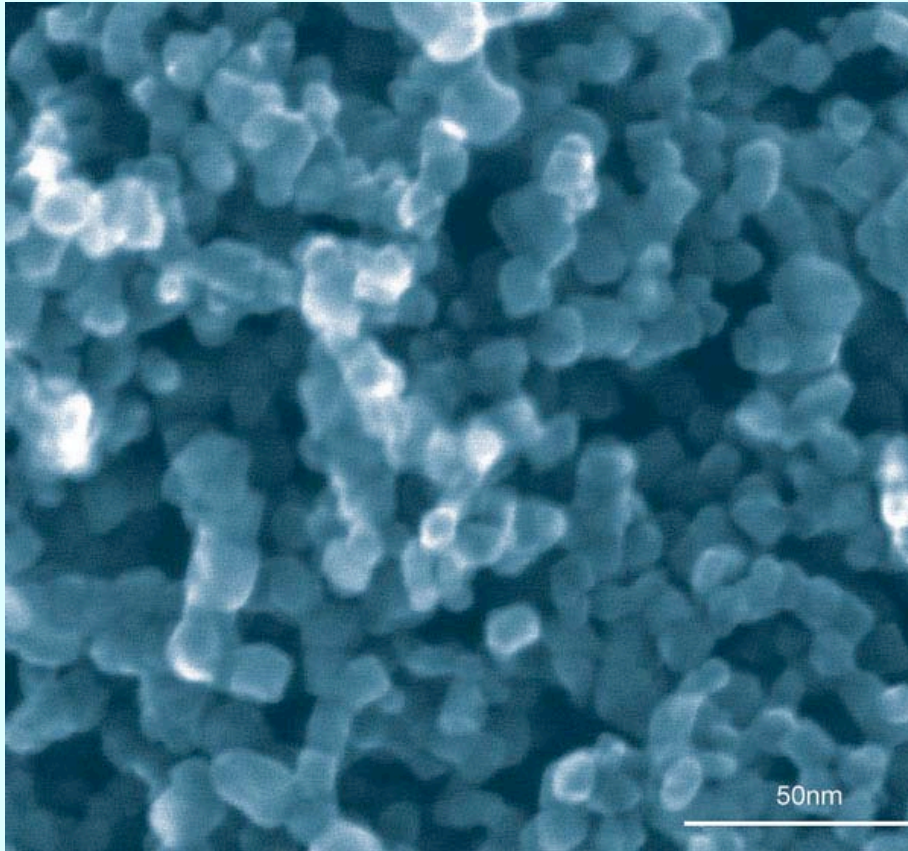


Cyclic voltammogram of the N621 complex anchored onto a TiO_2 electrode (black line), exposed to the solutions of HgCl_2 (red dashed dotted line) and $\text{Pb}(\text{ClO}_4)_2$ (blue dashed line)



Molecular structure of the **N719**-HgCl₂ complex showing the asymmetric unit which contains one mercury atom, and two Cl⁻ anions

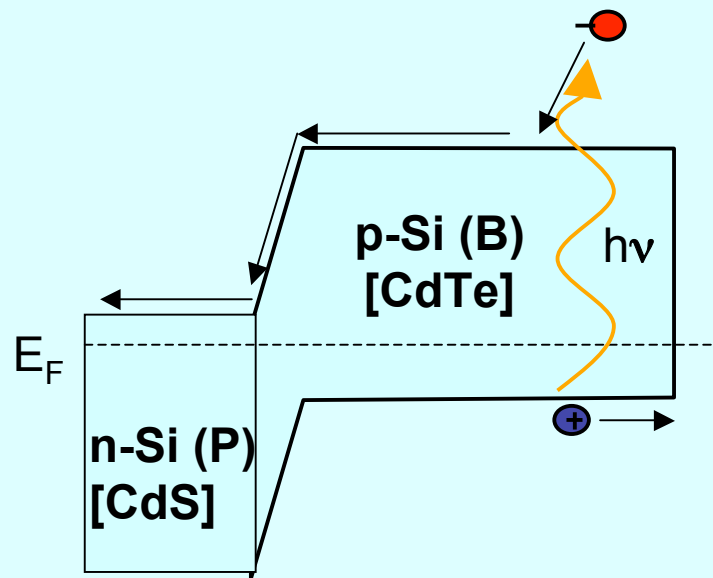




Outline

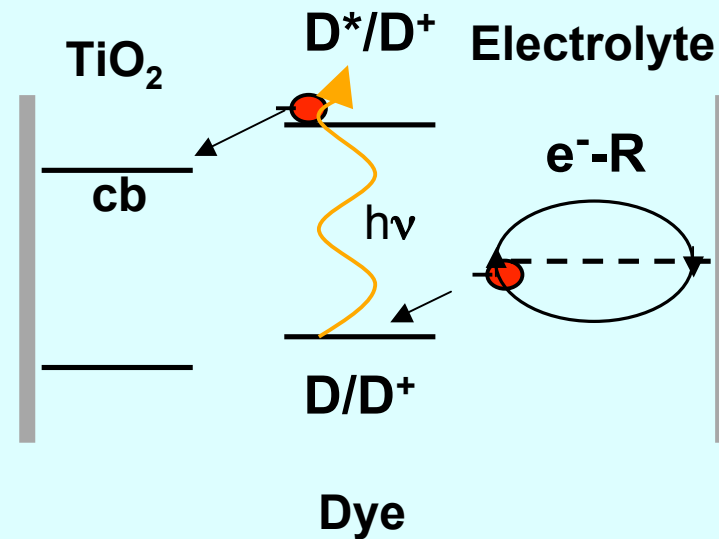
- Mesoscopic junctions, interfacial and cross surface charge transfer
- **Photoinduced charge separation**
- Photogalvanic generation of electricity from sunlight

Silicon Photovoltaic Cells

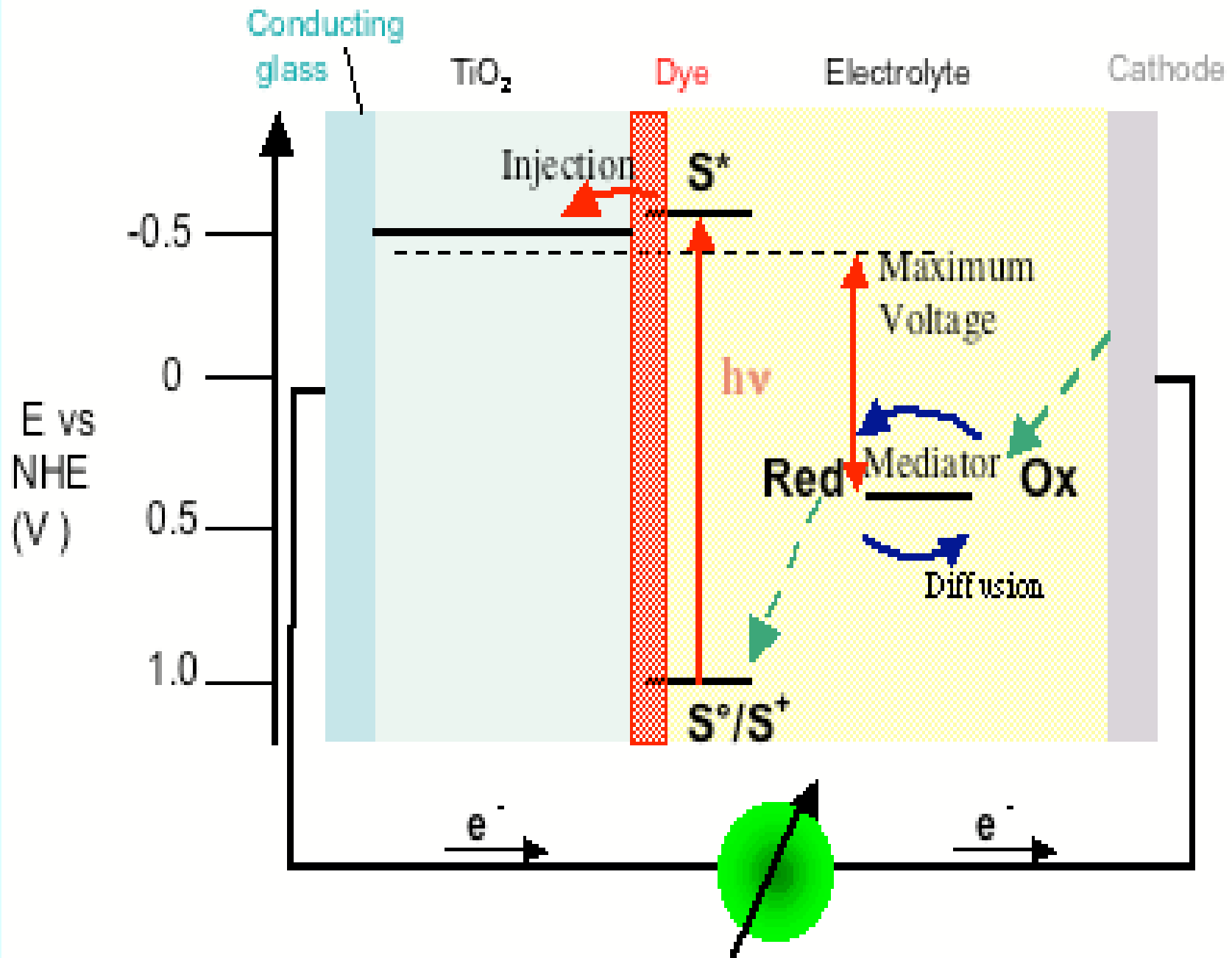


Charge separation by electric field within a p- and n-doped semiconductor material (Si, II-VI, a-Si: H)

Dye Solar Cells



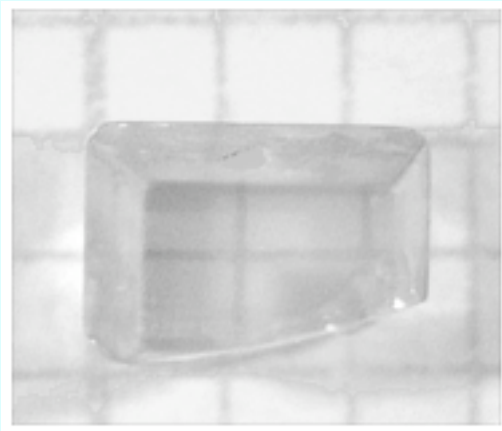
Charge separation by kinetic competition like in photosynthesis



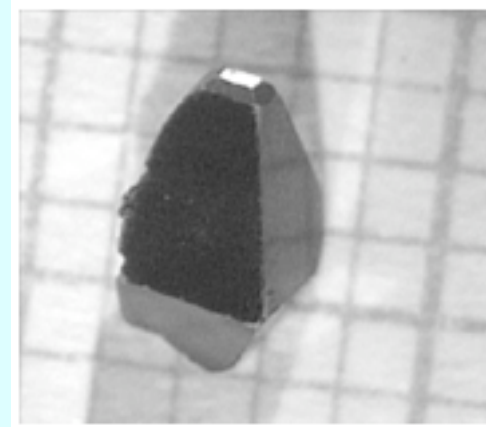
The two dilemmas of light harvesting by surface immobilized molecular absorbers

- 1. A monolayer of dye on a flat surface absorbs at most a few percent of light because it occupies an area that is much larger than its optical cross section**
- 2. Compact semiconductor films need to be n-doped to conduct electrons. Energy transfer quenching of the excited sensitizer by the electrons in the semiconductor leads to conversion of light to heat reducing photovoltaic conversion efficiency.**

Anatase crystals



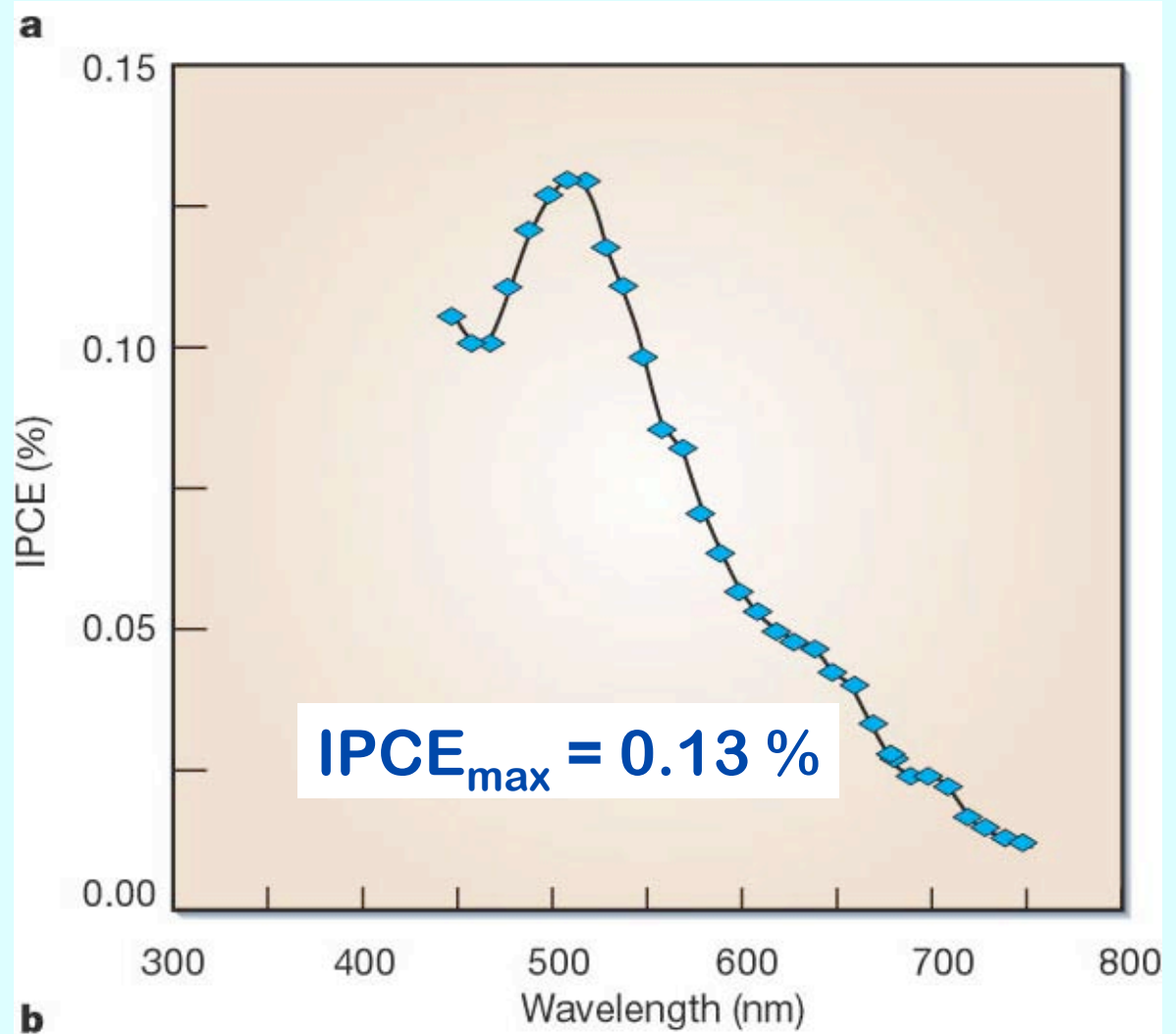
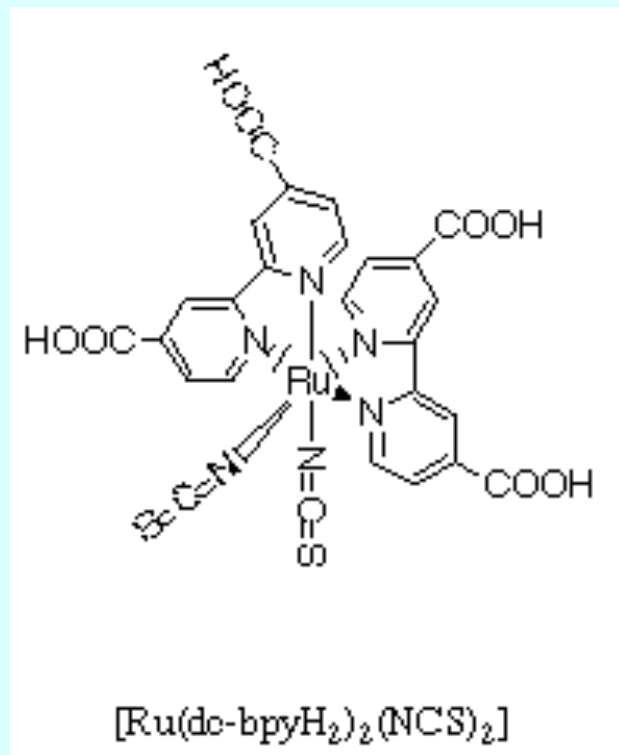
- Undoped crystal, (001) surface



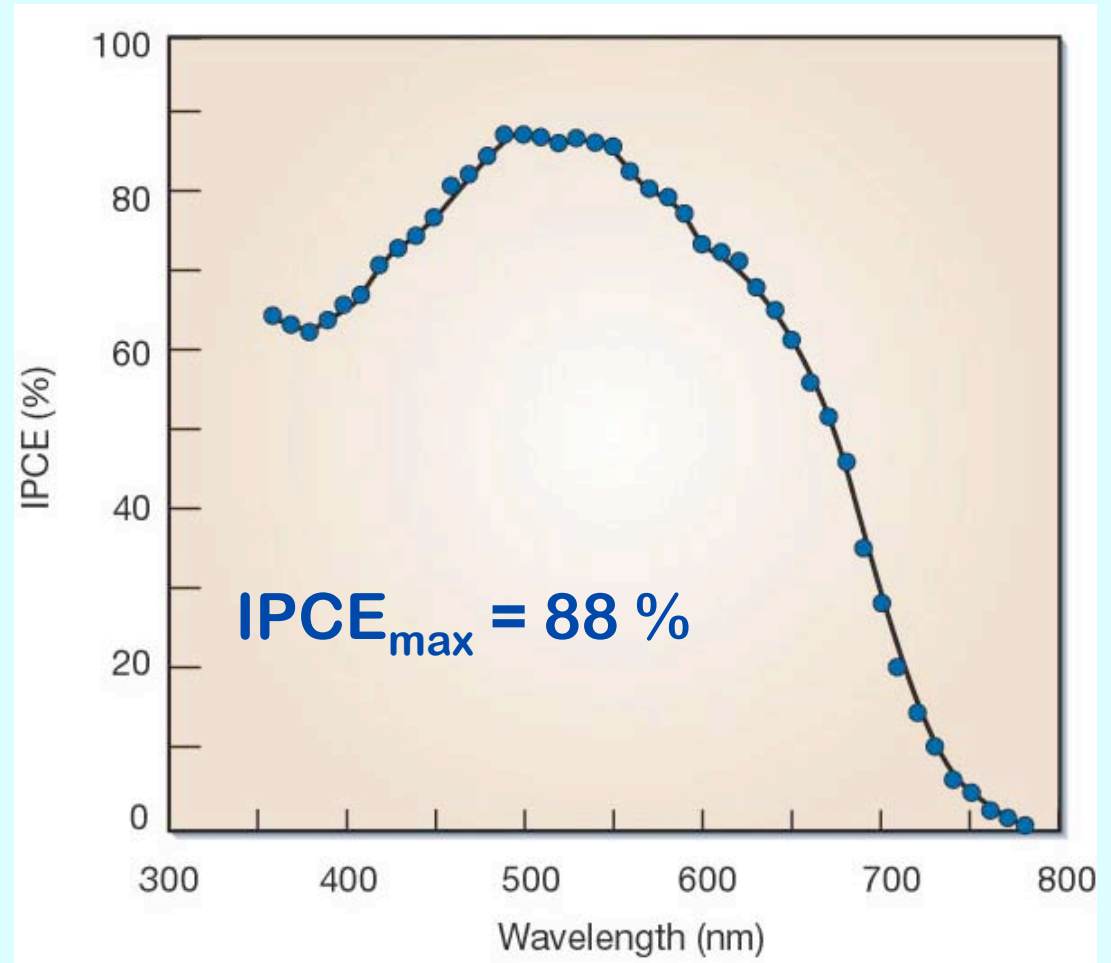
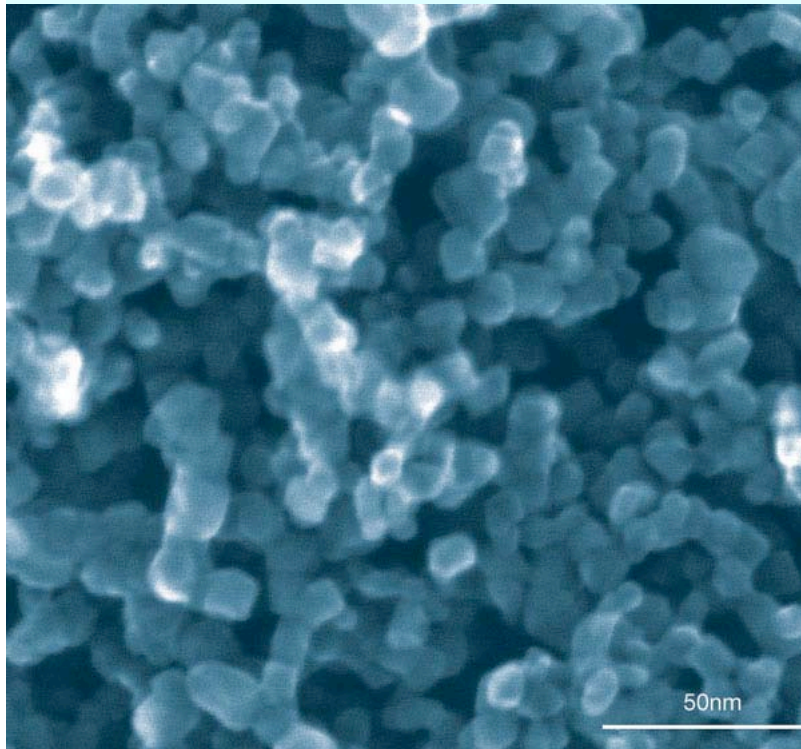
- Doped crystal, (101) surface

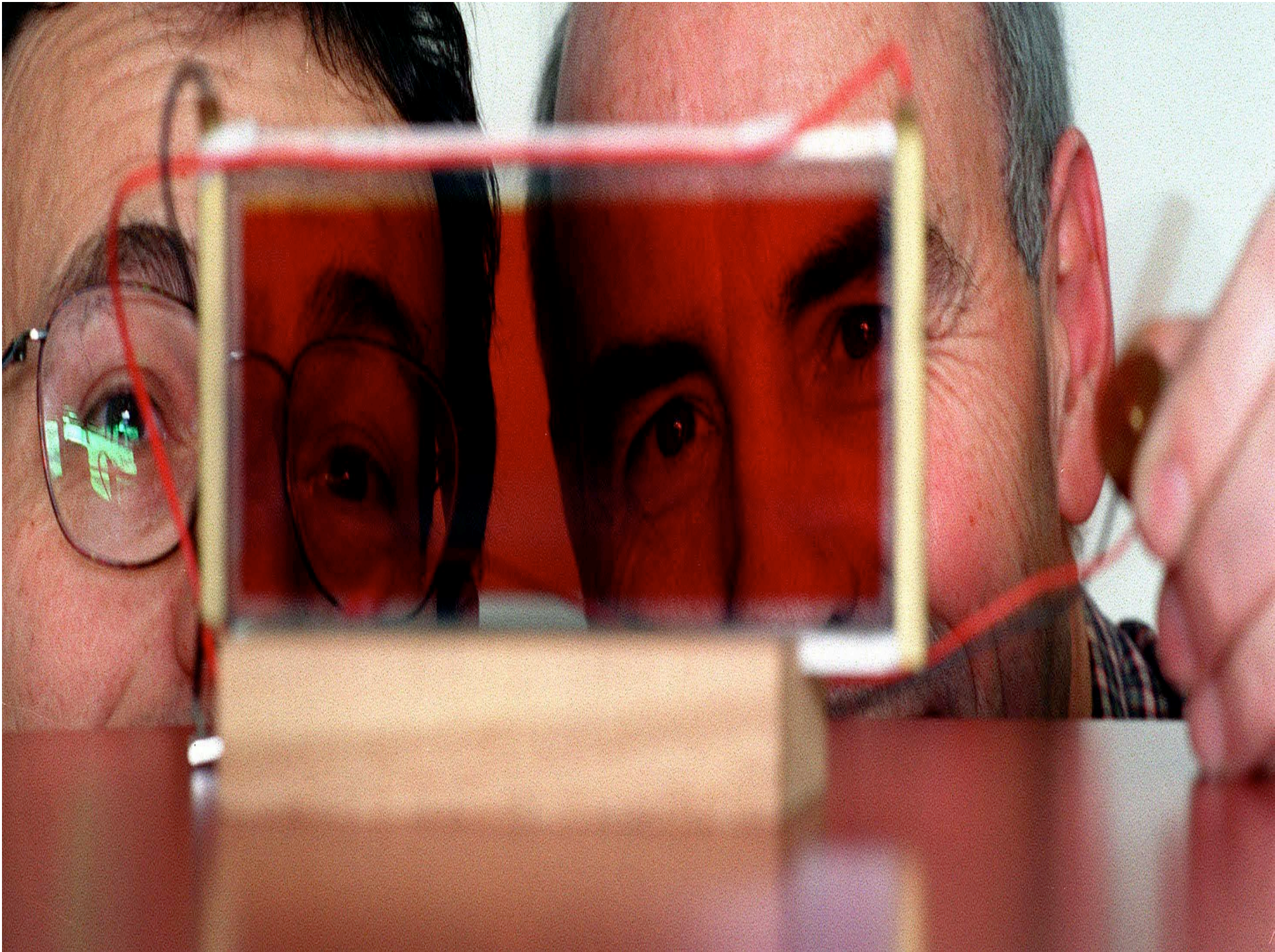
A. Vittadini, A. Selloni, F. Rotzinger and M. Grätzel Phys. Rev. Lett. 81, 2954 (1998)

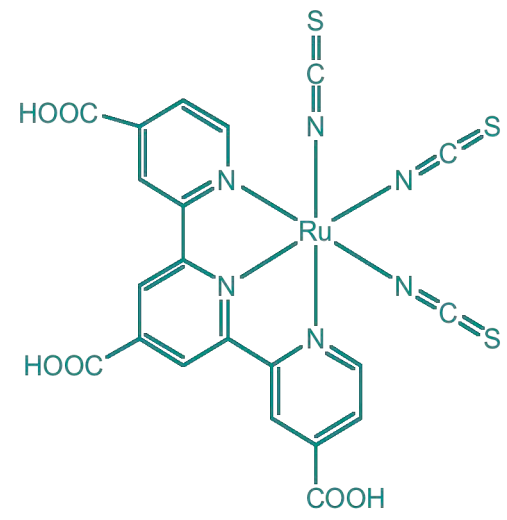
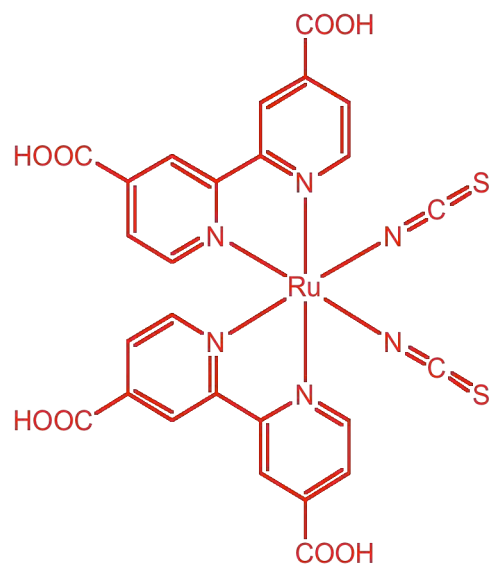
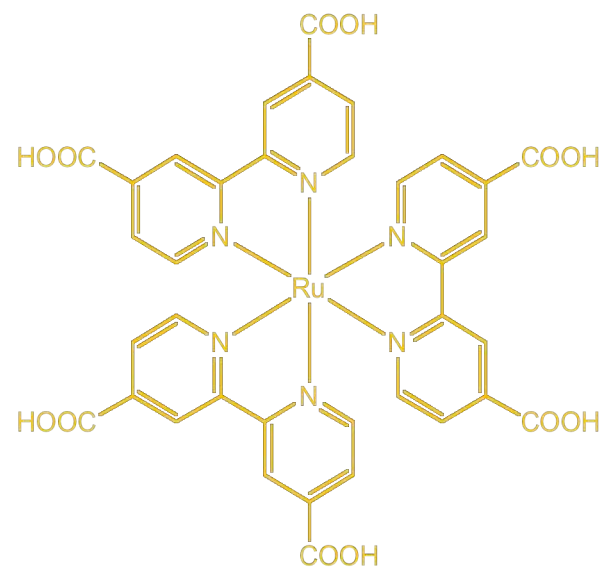
Incident photon to electron conversion efficiency (IPCE) of a dye-sensitized TiO₂ (101) single crystal PEC solar cell



Incident photon to current conversion efficiency of a dye-sensitized solar cell based on a mesoscopic TiO_2 electrode

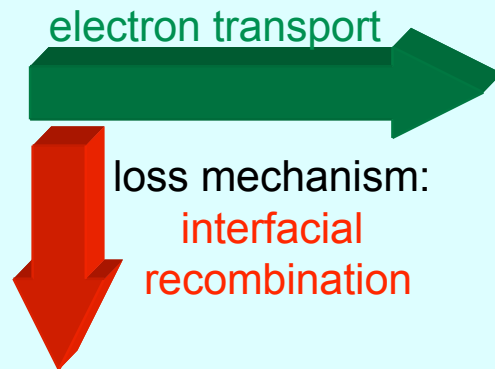
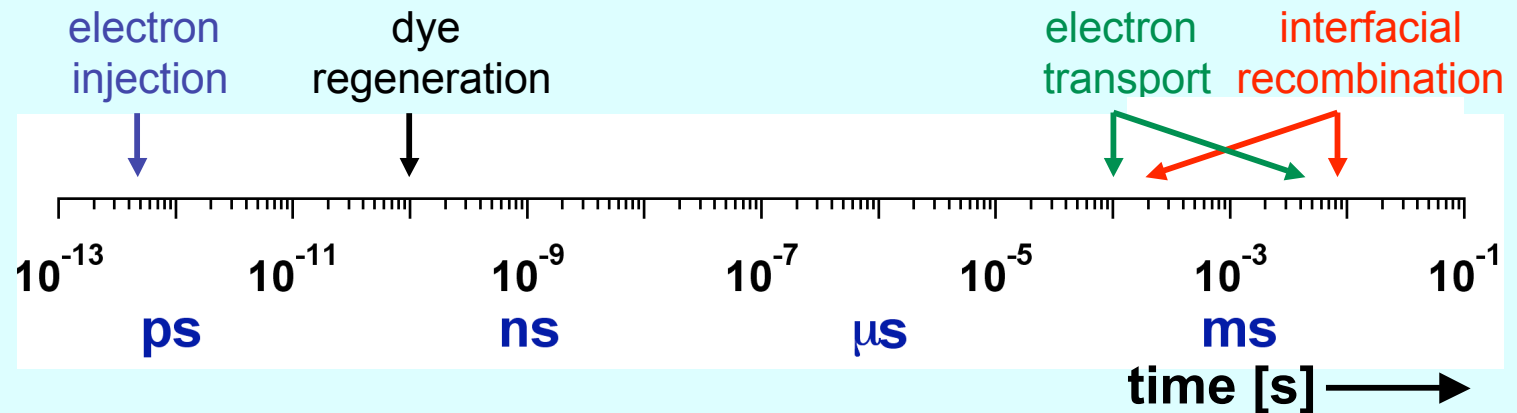






**Dye sensitized nanocrystals
show quantitative conversion
of the photons into electric
current**

Dynamic Competition



Competition ⇒

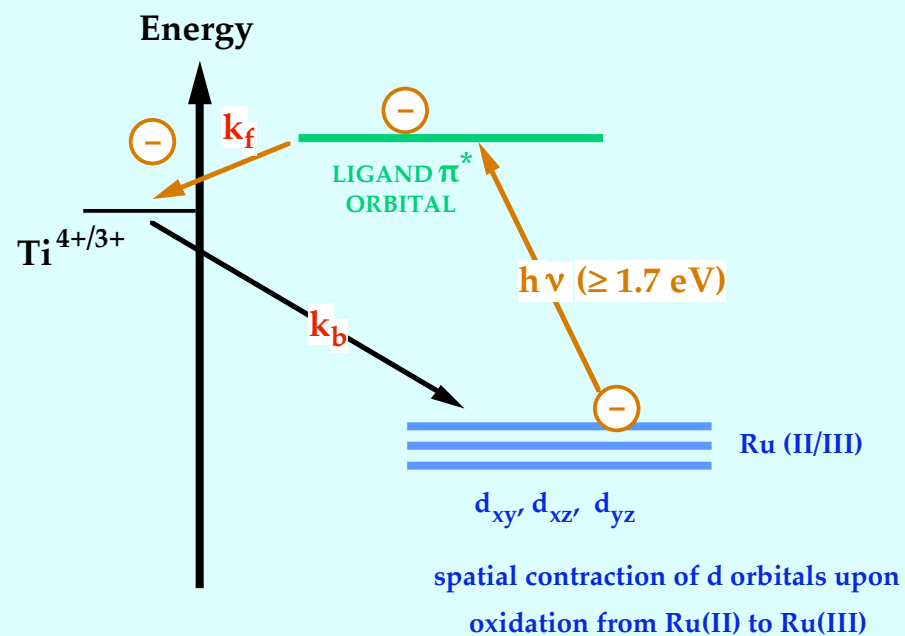
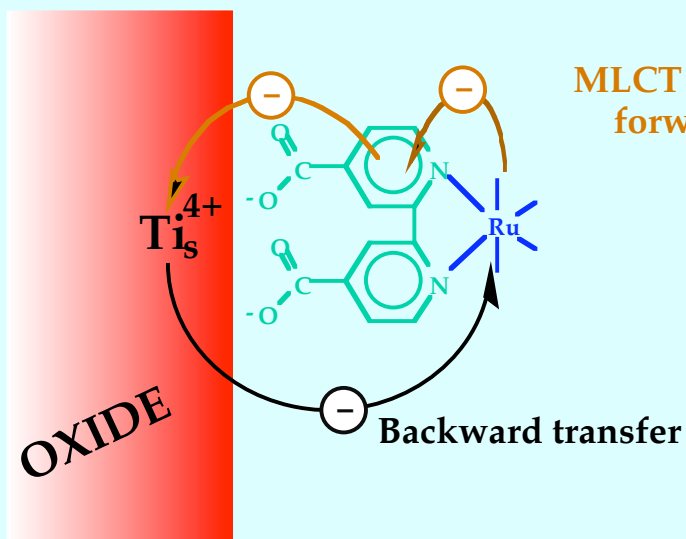
Electron diffusion length

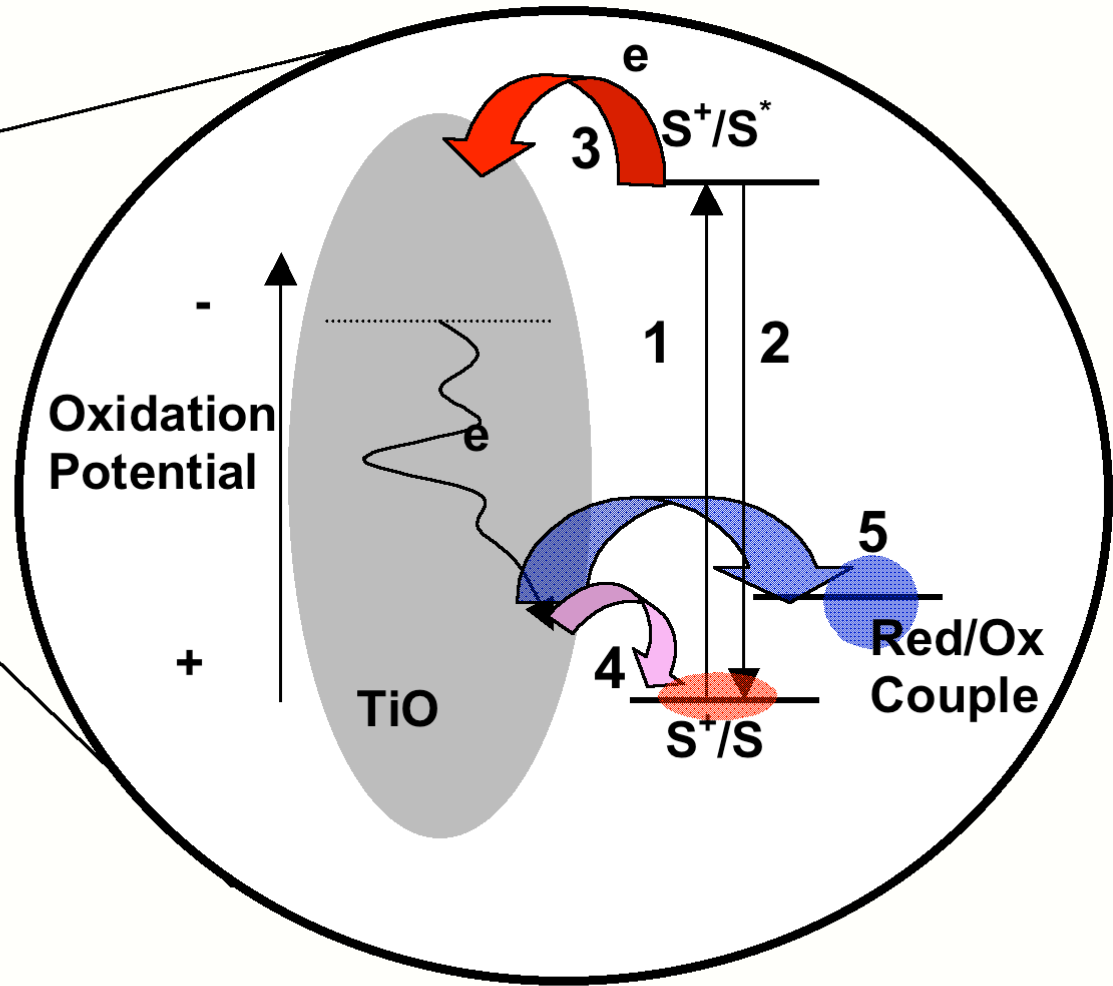
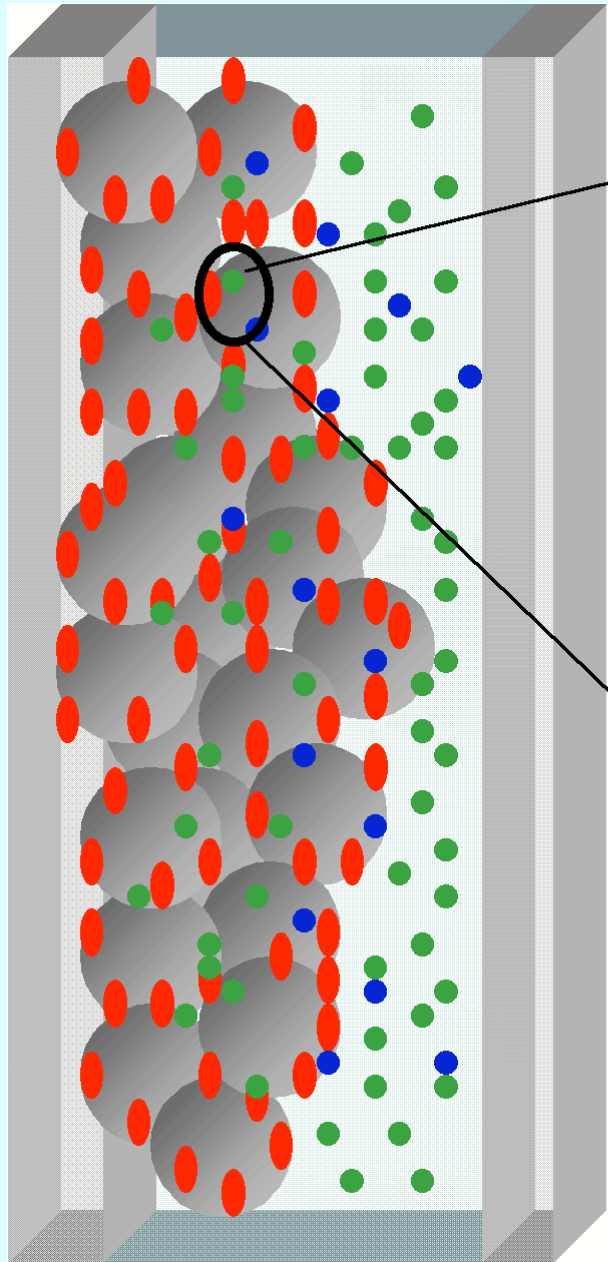
$$L_n = \sqrt{D_n \cdot \tau_n}$$

τ_n : electron lifetime

D_n : electron diffusion coefficient

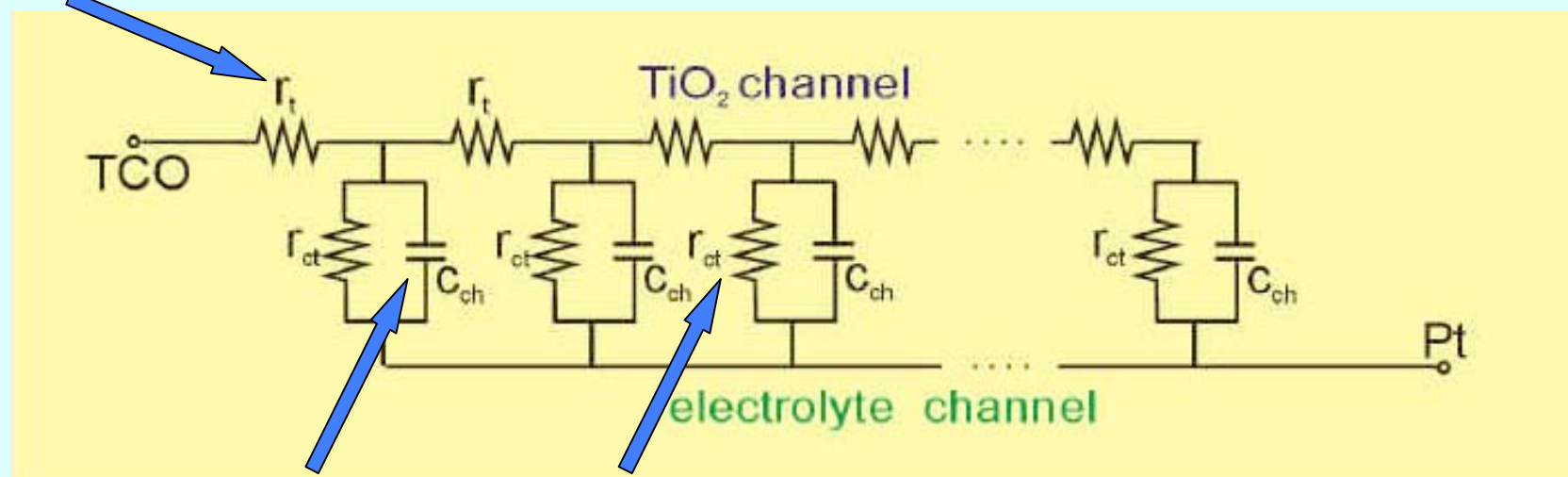
Photo Induced Heterogeneous Electron Transfer Cycle



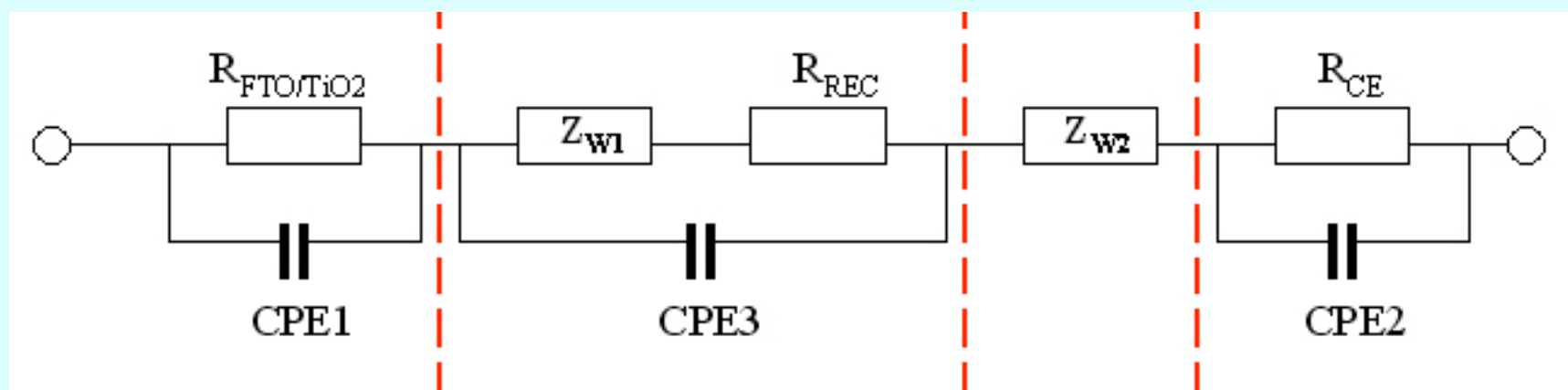


Finite length transmission model (Bisquert)

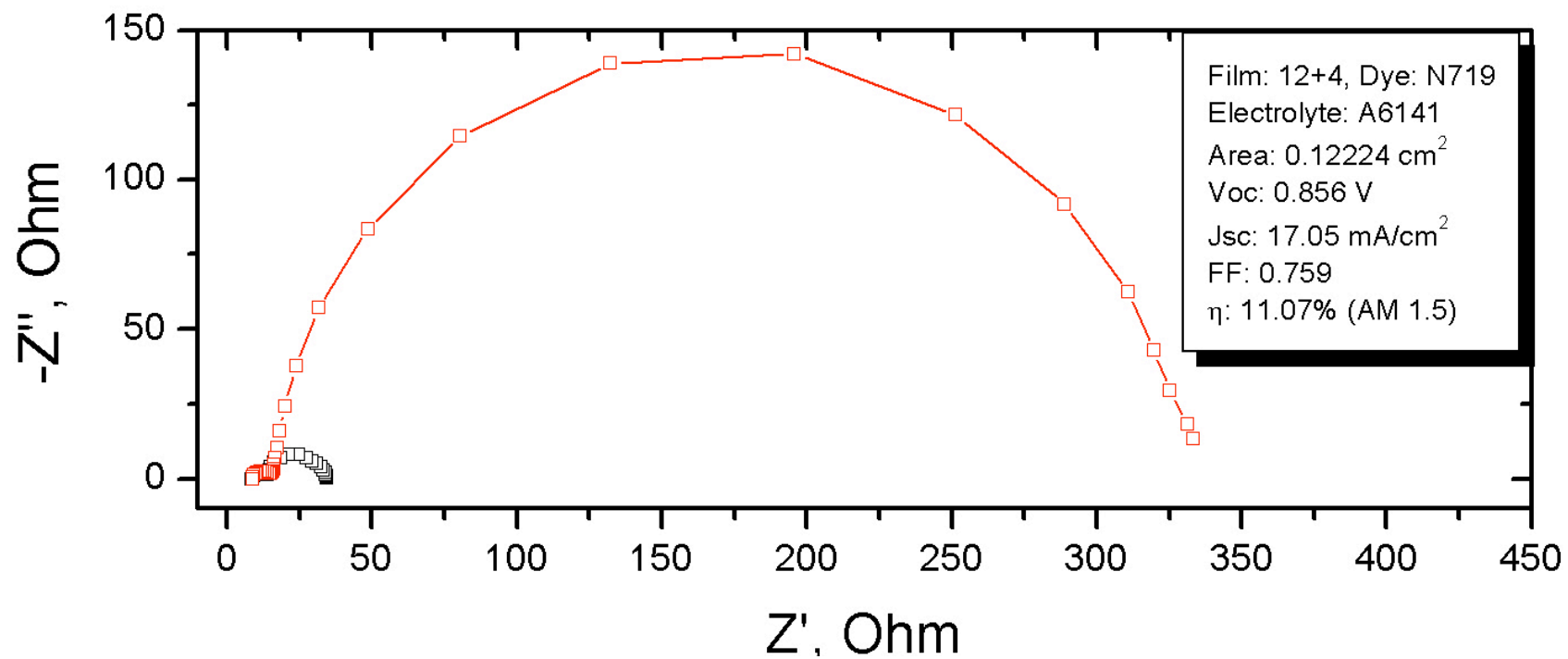
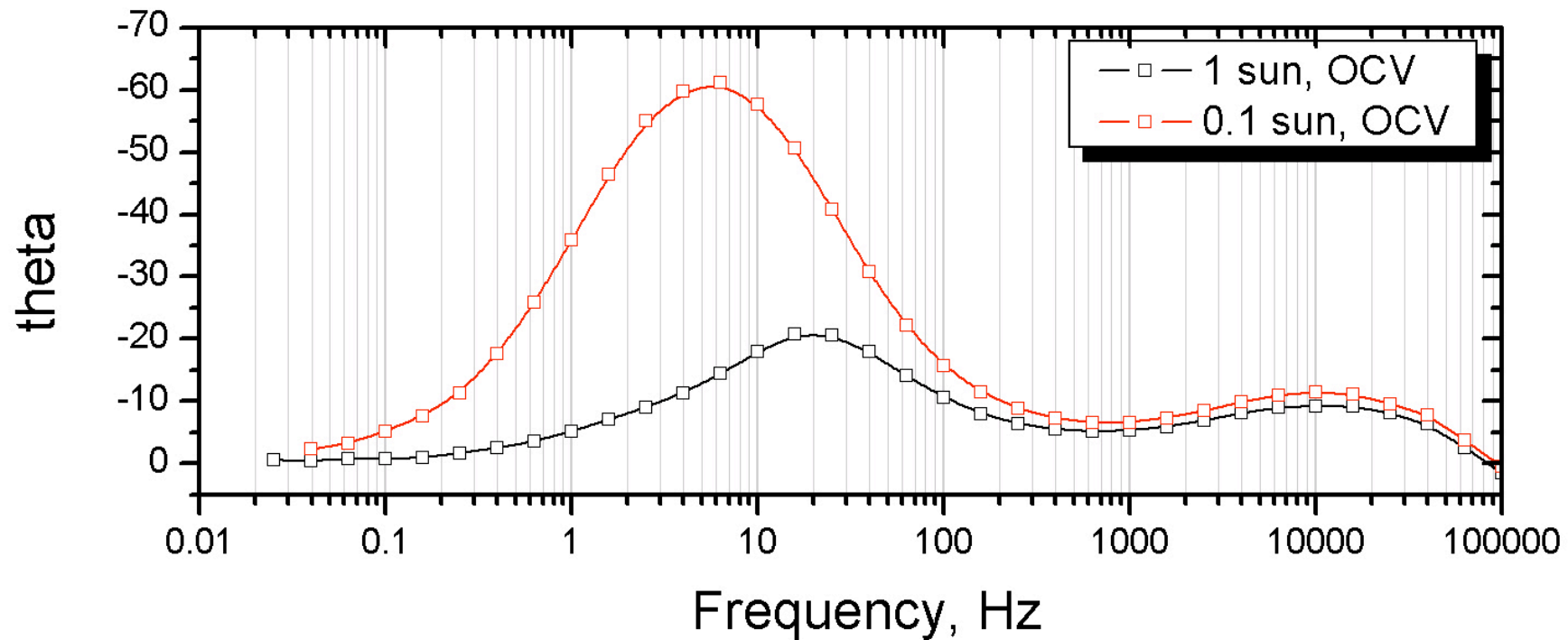
Transport

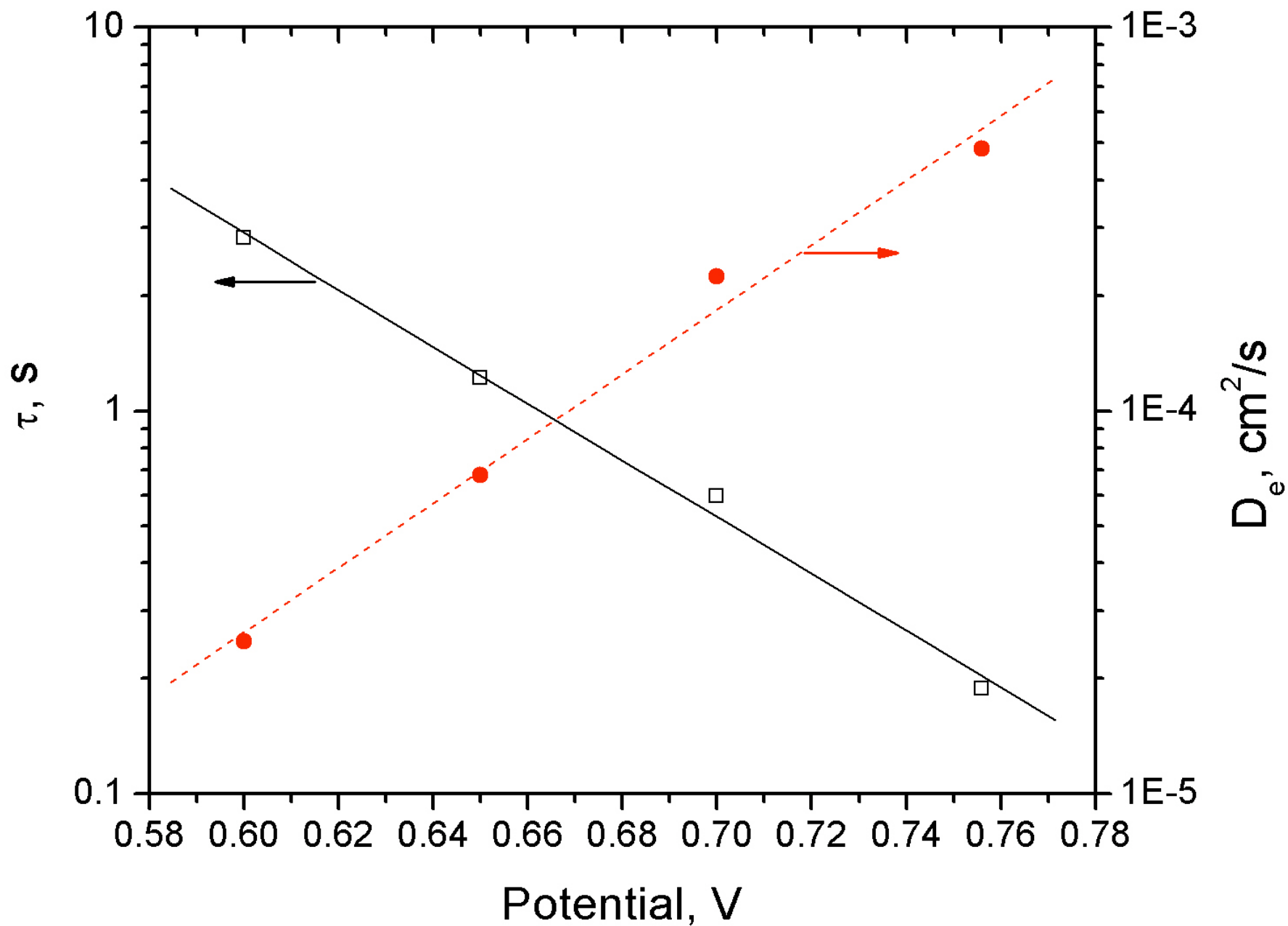


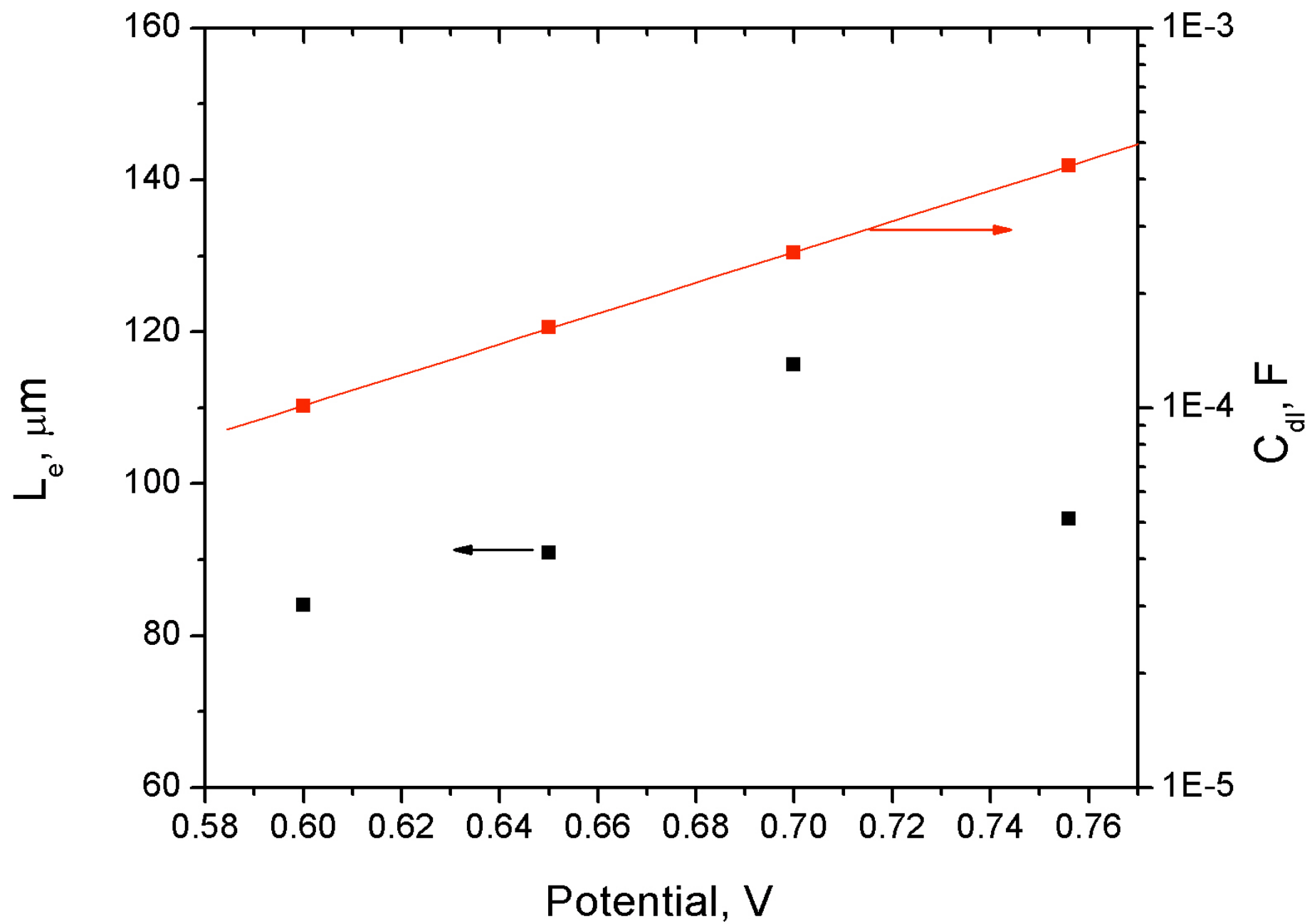
Chemical Capacitance Recombination



Q Wang, J. Moser and M. Graetzel J.Phys. Chem B in press

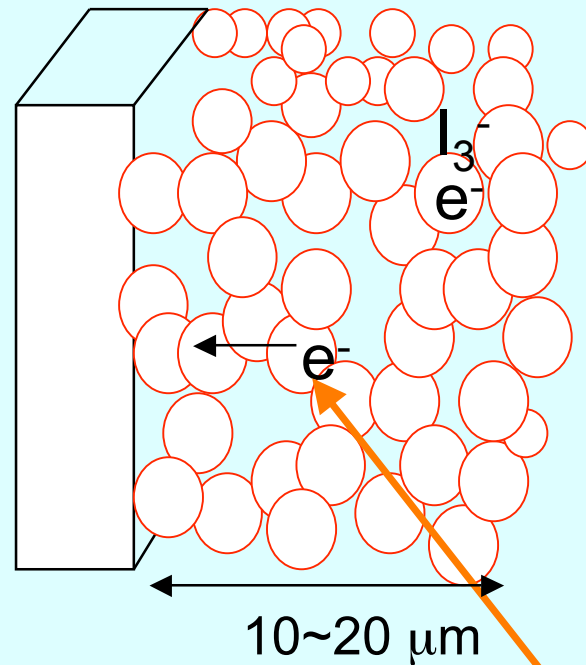






Electron Transport: Diffusion and Electron Lifetime

SnO₂:F TiO₂/Electrolyte



$$L = \sqrt{D\tau}$$

Charge recombination
 $2 e^- + I_3^- = 3 I^-$

Electrons should travel to the SnO₂ before charge recombination occurs

Diffusion length should exceed the thickness of the mesoscopic TiO₂ film

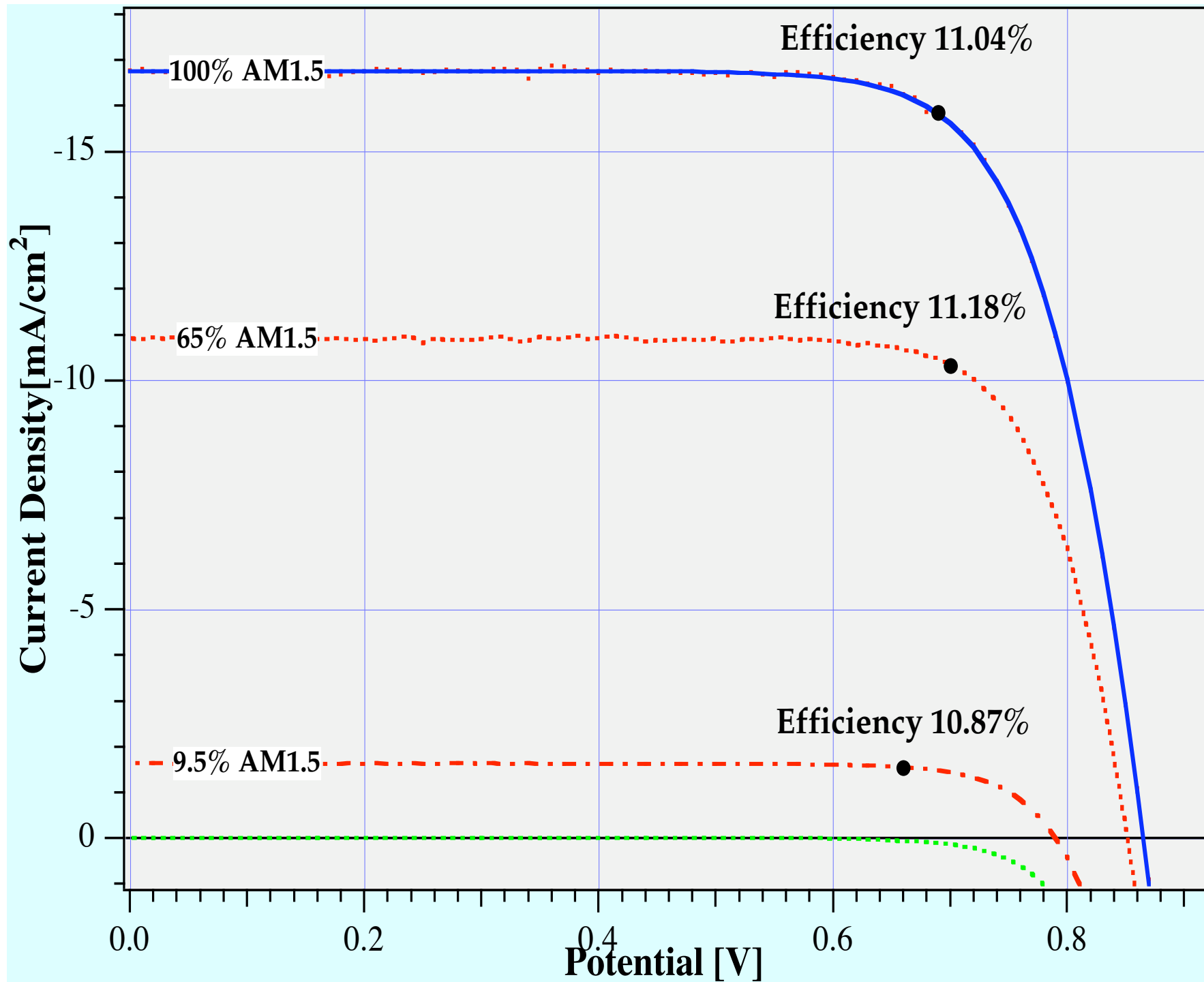
Increasing the injection and lowering the recombination rates is critical for maximizing the open circuit voltage of the cell !

$$V_{oc} = (nRT/F) \ln(K\Phi / (k_1[S^+] + k_2[D^+]))$$

$K\Phi$: charge carrier photo-generation rate

k_1, k_2 : recombination rate constants

n : ideality factor of the junction



STABILITY

Requirements for outdoor use according to international PV standards applied to single crystal silicon but so far not to thin film PV cells

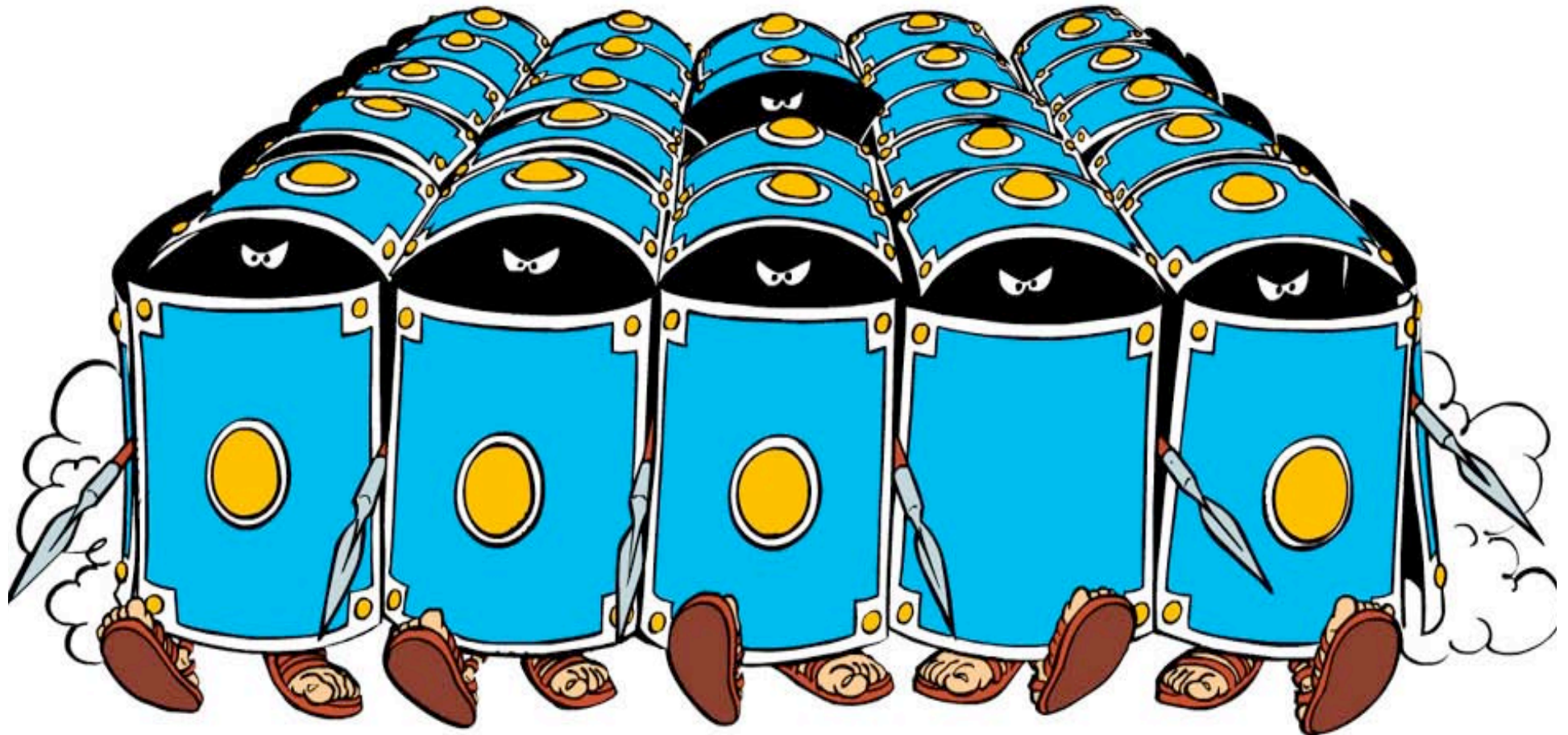
UV plus heat (55-60 C): 1000 hours

Accelerated thermal test at 85 C: 1000 h

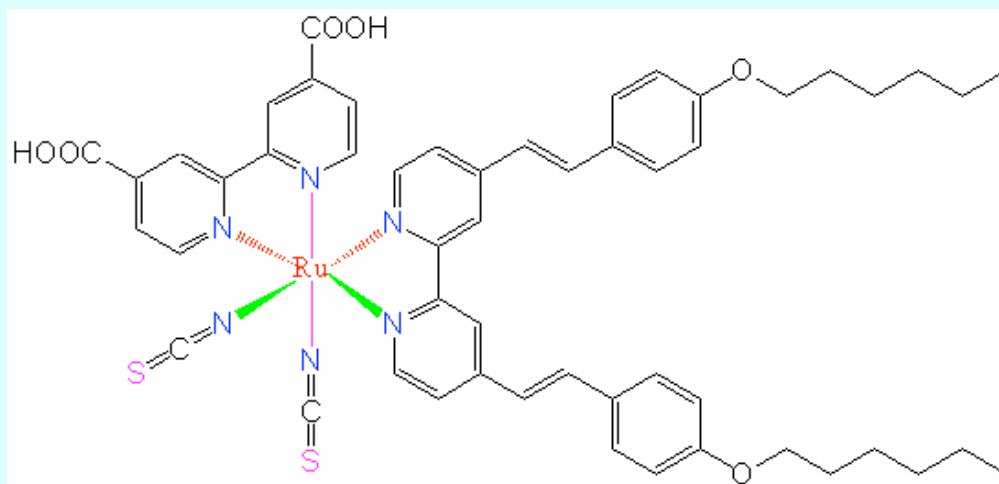
Humidity test and temperature cycling (sealing issues)

Self-assembly of stable and well defined monomolecular layers of sensitizer at the interface provides long term photovoltaic stability and high conversion efficiency

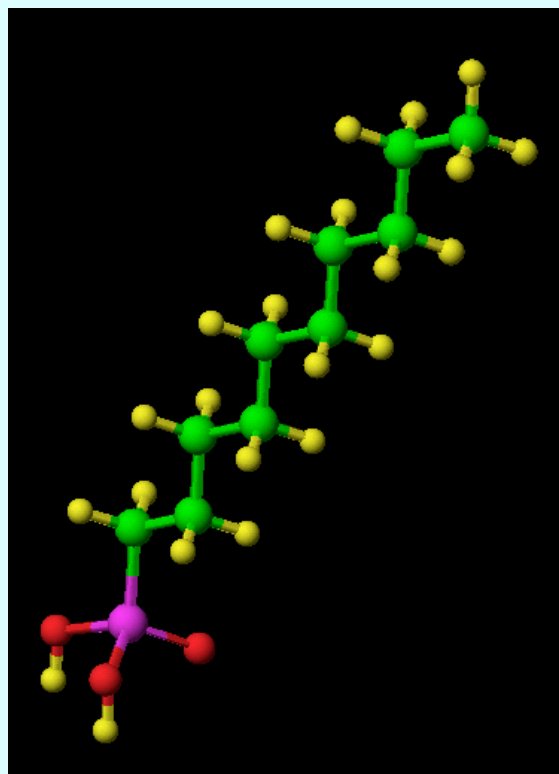
Interface Engineering in Dye-Sensitised Solar Cells



K-19



Decylphosphonate



Photoanode: 8+5

***ROBUST
Electrolyte***

PMII: 0.8 M

I₂: 0.15 M

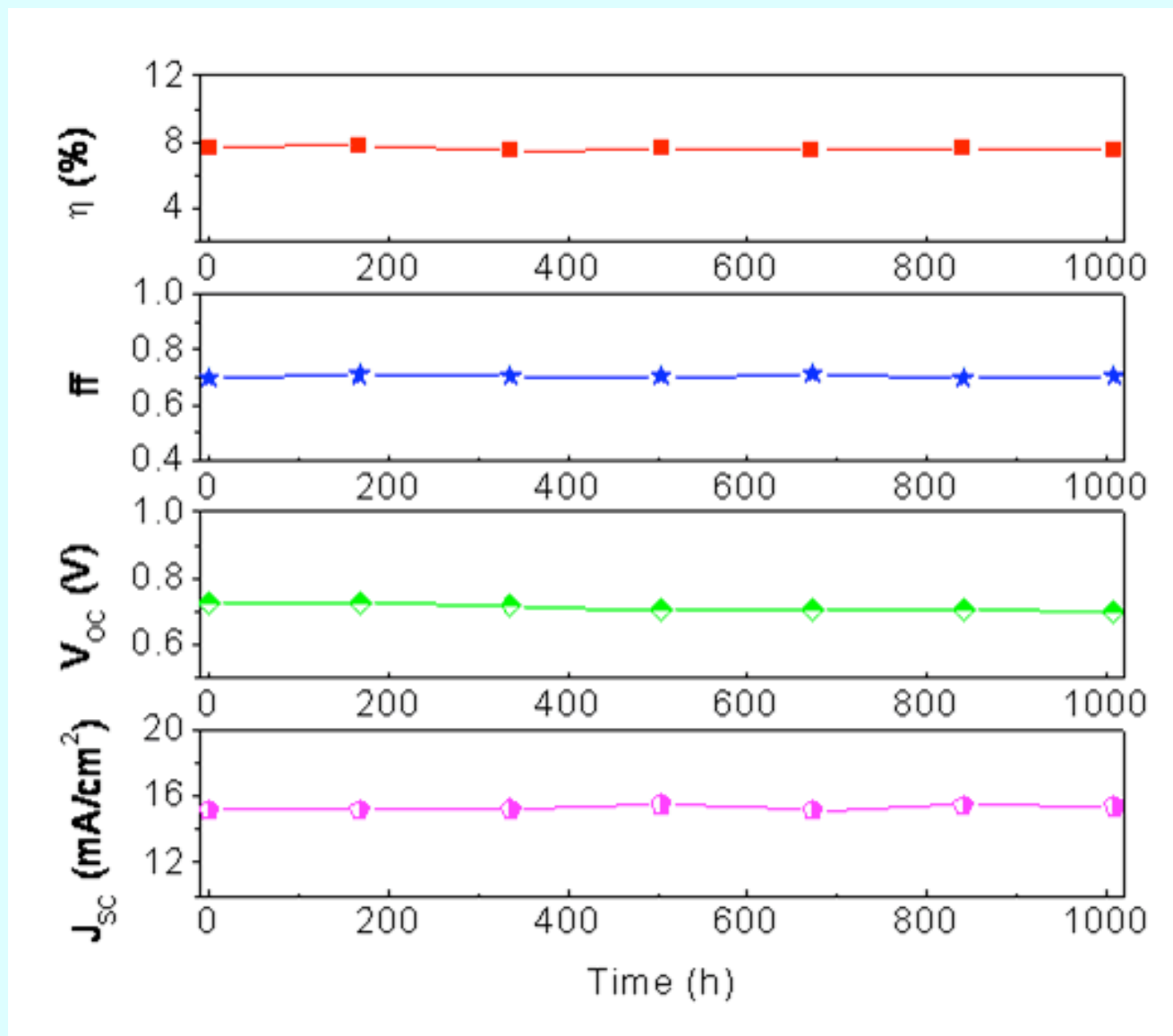
NMBI: 0.5 M

0.1 M GSCN

MPN solvent

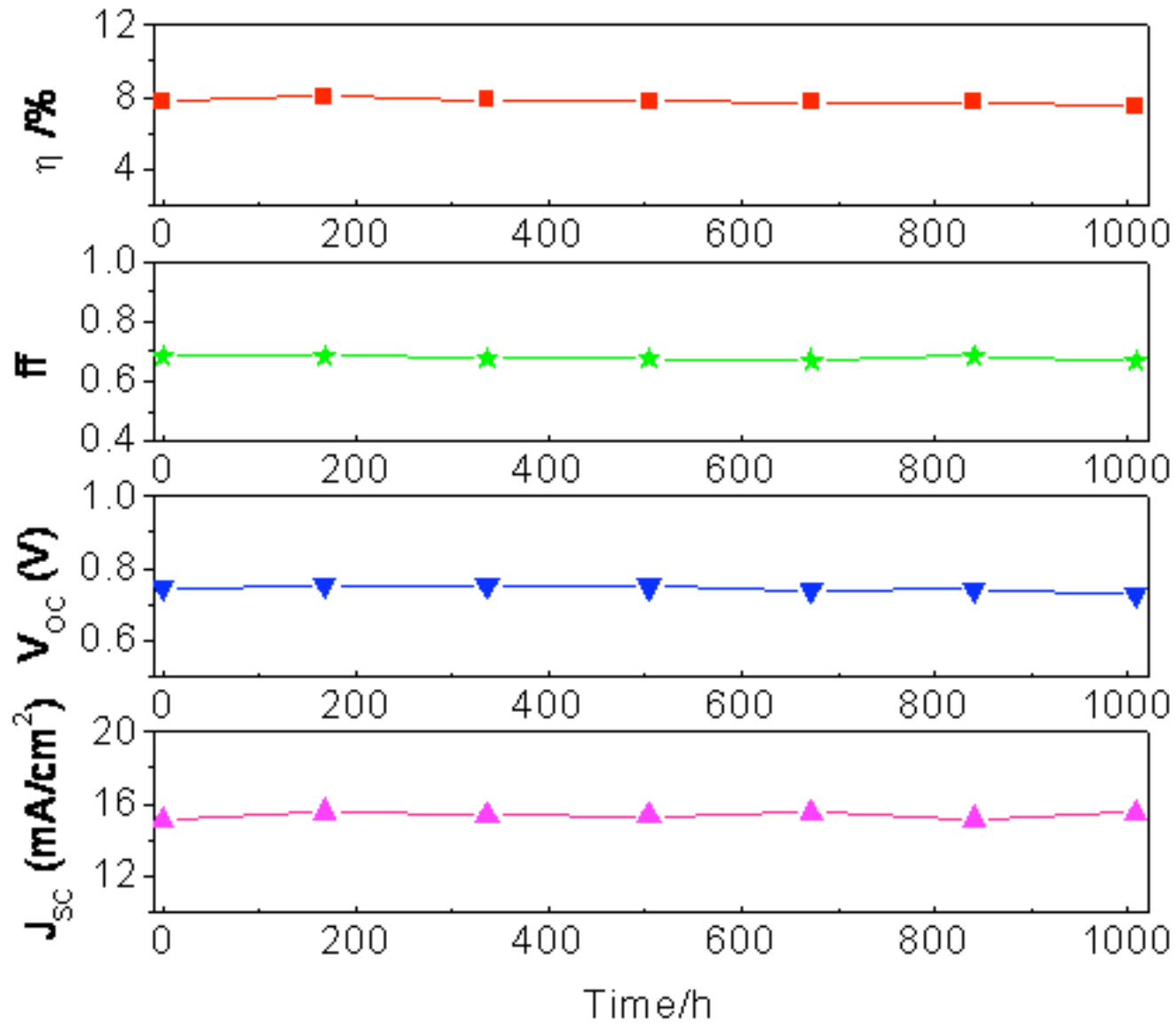
Efficiency: > 8.0%

80 °C evolution of device parameters in the dark



Wang, P.; Klein, C.; Humphry-Baker, R.; Zakeeruddin, S. M.; Grätzel, M. *Appl. Phys. Lett.* 2005, 86, 123508.

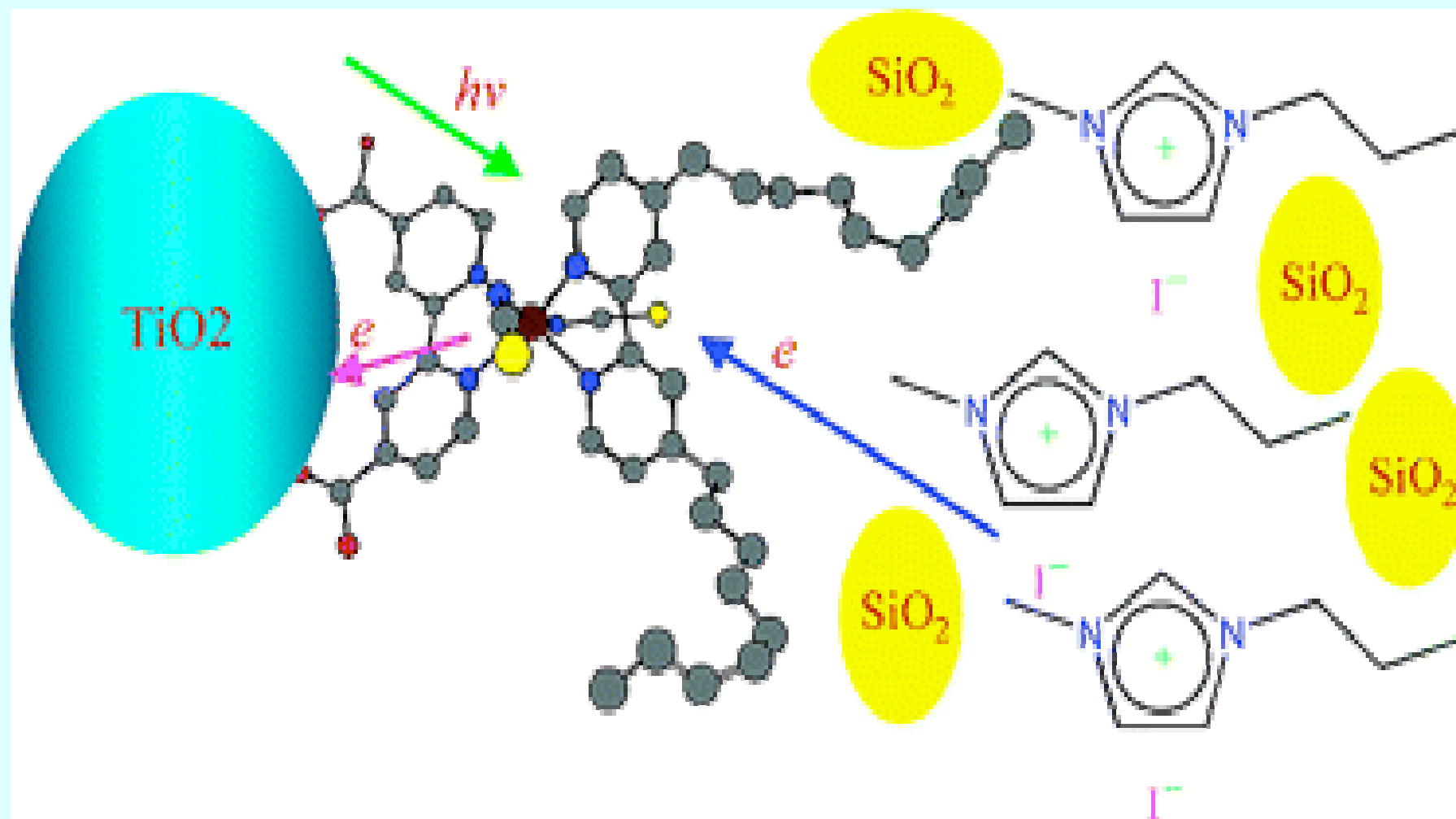
60 °C evolution of device parameters under one sun soaking



SOLVENT-FREE SYSTEMS

**SOLID (POLYMER)ELECTROLYTES,
SOLIDIFIED IONIC LIQUIDS
HOLE CONDUCTORS**

Ionic solid electrolytes

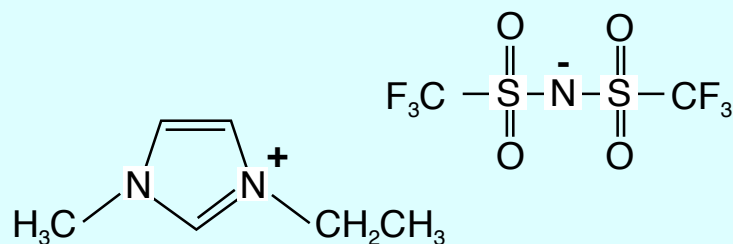


P. Wang, S.M. Zakeeruddin, P. Comte, I. Exnar, and M. Grätzel
Peng Wang et al *J. Am. Chem. Soc* 2003, 125, 1166-1167

ION-GEL Electrolyte (NEDO)

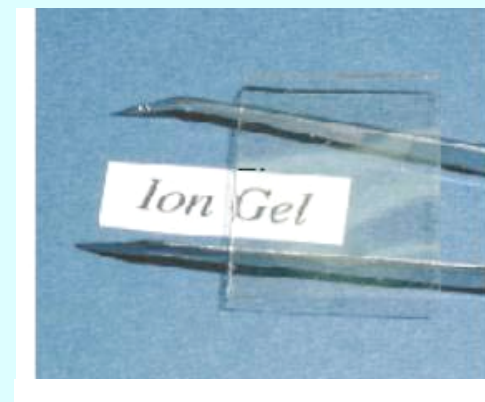
Features of Ionic Liquids

- Consists of only ions
- Liquid under wide temp. range ex. -10°C to 400°C
- non volatile
- Chemically stable and non combustible
- High electronic conductivity



1-Ethyl-3-methylimidazolium - Bis(trifluoromethylsulfonyl)
Amide

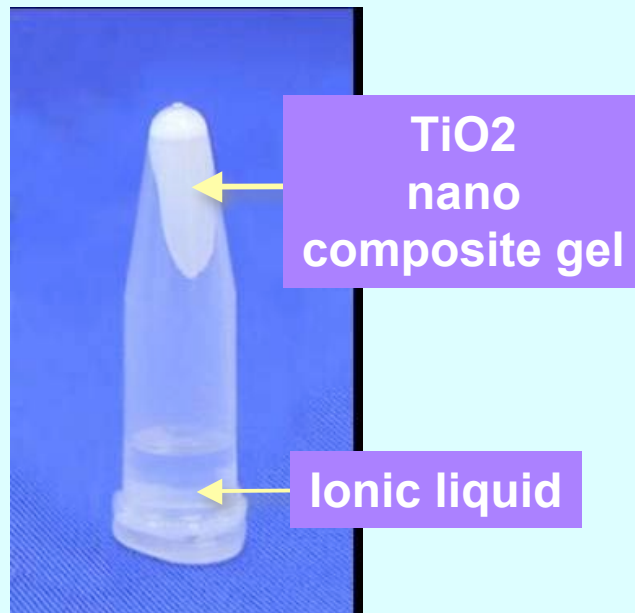
EMIm-TFSA



Nano composite Ion Gel

References

- (1) T.Fukushima, A.Kosaka, Y.Ishimura, T.Yamamoto, T.Takigawa, N.Ishii and T.Aida, *Science*, 27(2003)2072.
- (2) P.Wang, S.M.Zakeeruddin, P.Comte, I.Exnar and M.Graetzel, *J.Am.Chem.Soc.*, 125(2003)1166



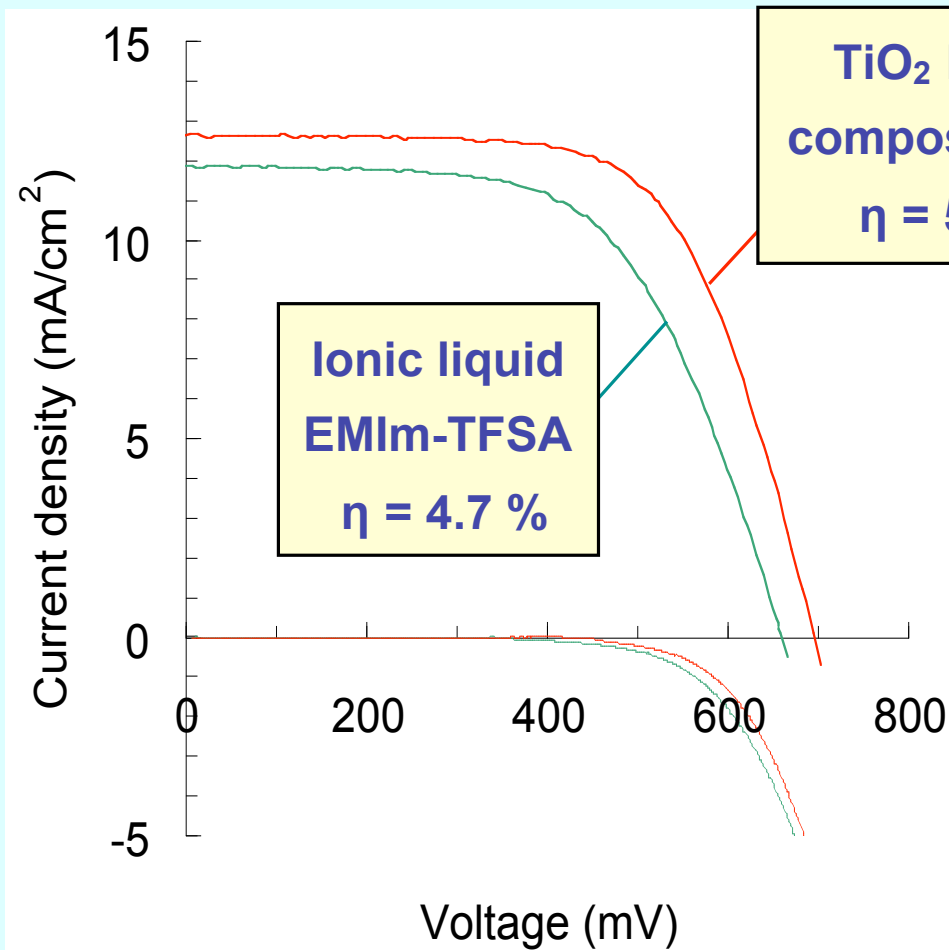
**EMIm-TFSA + TiO₂
(P25, 10 wt%)**

**Centrifugal separation
2000 G (6400 rpm) x 1hr**



I₂ content

Influence of Nano Particles



TiO₂ Nano composite gel
 $\eta = 5.7 \%$

Ionic liquid EMIm-TFSA
 $\eta = 4.7 \%$



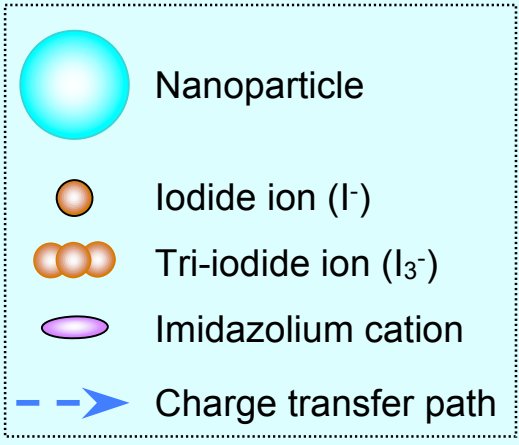
TiO₂ (P25); anatase, 28 nm

Table PV performance of nano composite

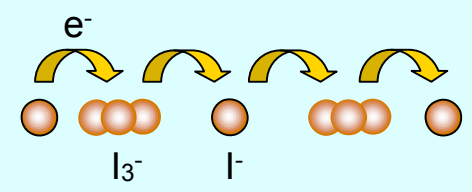
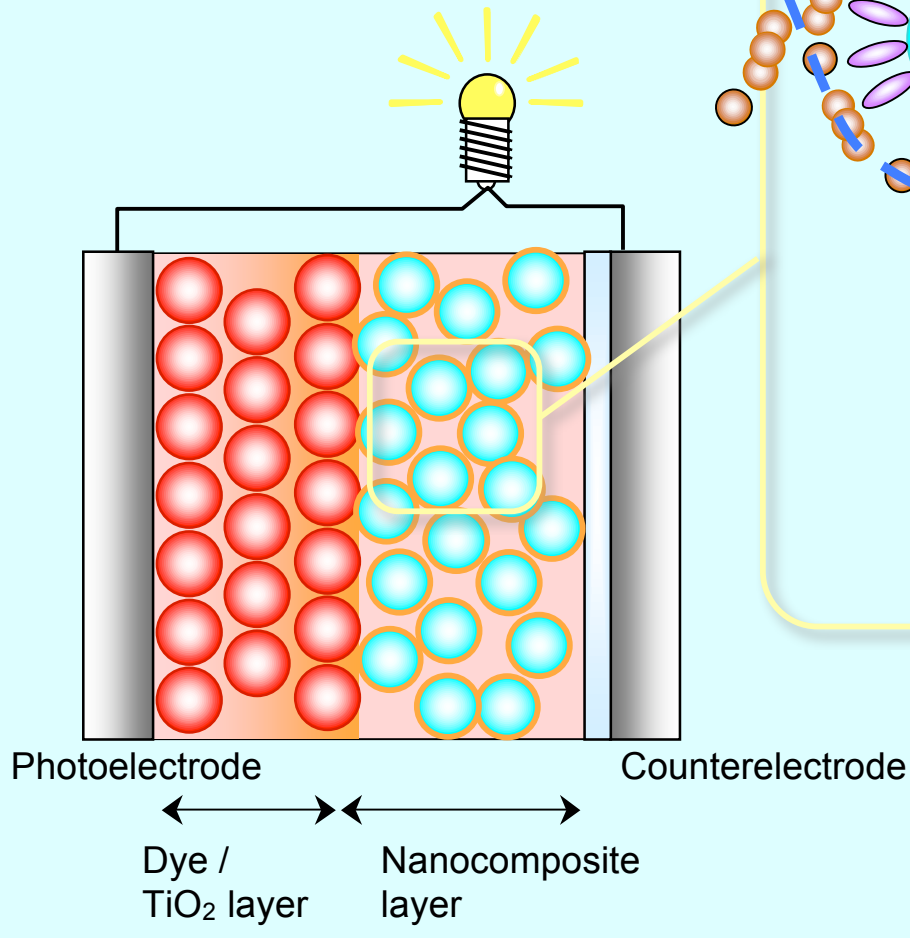
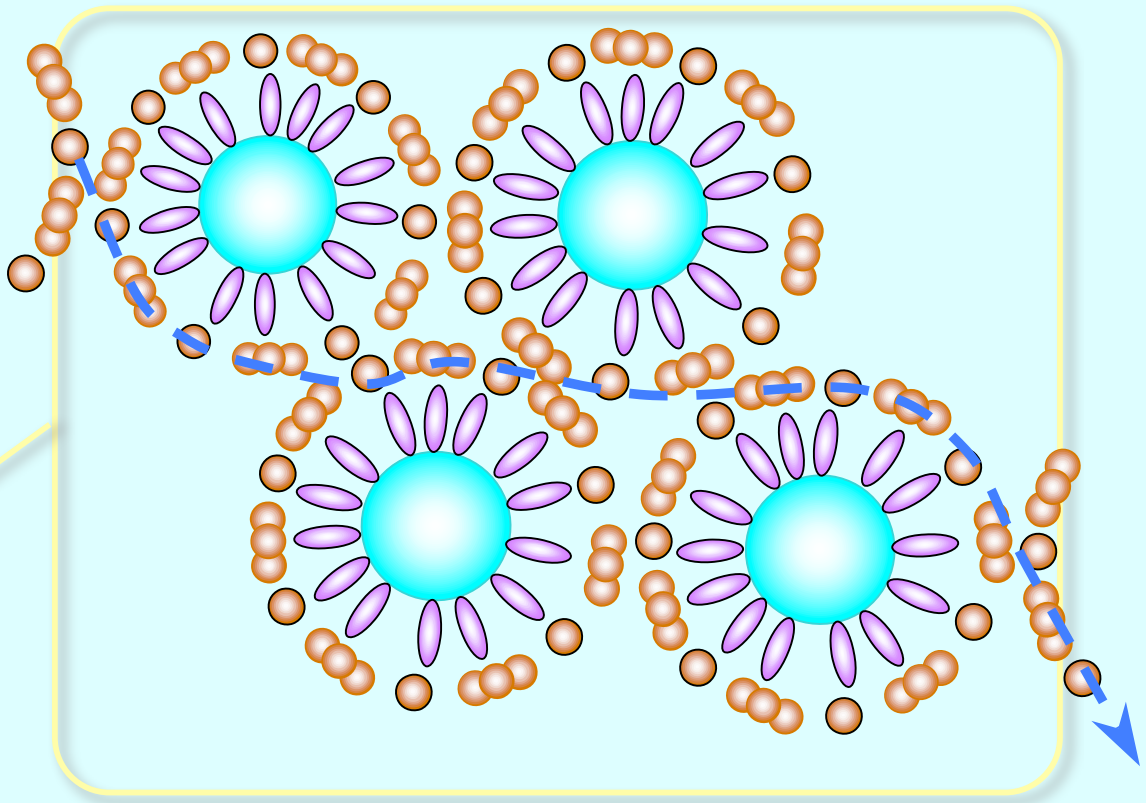
nano particles	ion gel cells J _{sc} (mA/cm ²)	V _{oc} (mV)	FF	η (%)
TiO₂	12.5	696	0.65	5.7
SiO₂	12.5	719	0.64	5.8
SnO₂	12.1	679	0.61	5.0
ITO	12.5	685	0.60	5.1
MW-CNT	12.0	706	0.57	4.8
without particles	11.8	661	0.60	4.7

Fig. I-V characteristics of nano composite ion gel cell and bare ionic liquid cell

Courtesy of Dr. Nobuo Tanabe Fujikura Ltd



Charge transportation in nano composite gel



Grotthus-like exchange mechanism

Latent gel electrolyte precursors for quasi-solid dye sensitized solar cells

Takehito Kato, Akio Okazaki and Shuzi Hayase* [Chem. Commun. 2005,363-365](#)

Received (in Cambridge, UK) 12th August 2004, Accepted 23rd September 2004

First published as an Advance Article on the web 29th November 2004

DOI: 10.1039/b412462f

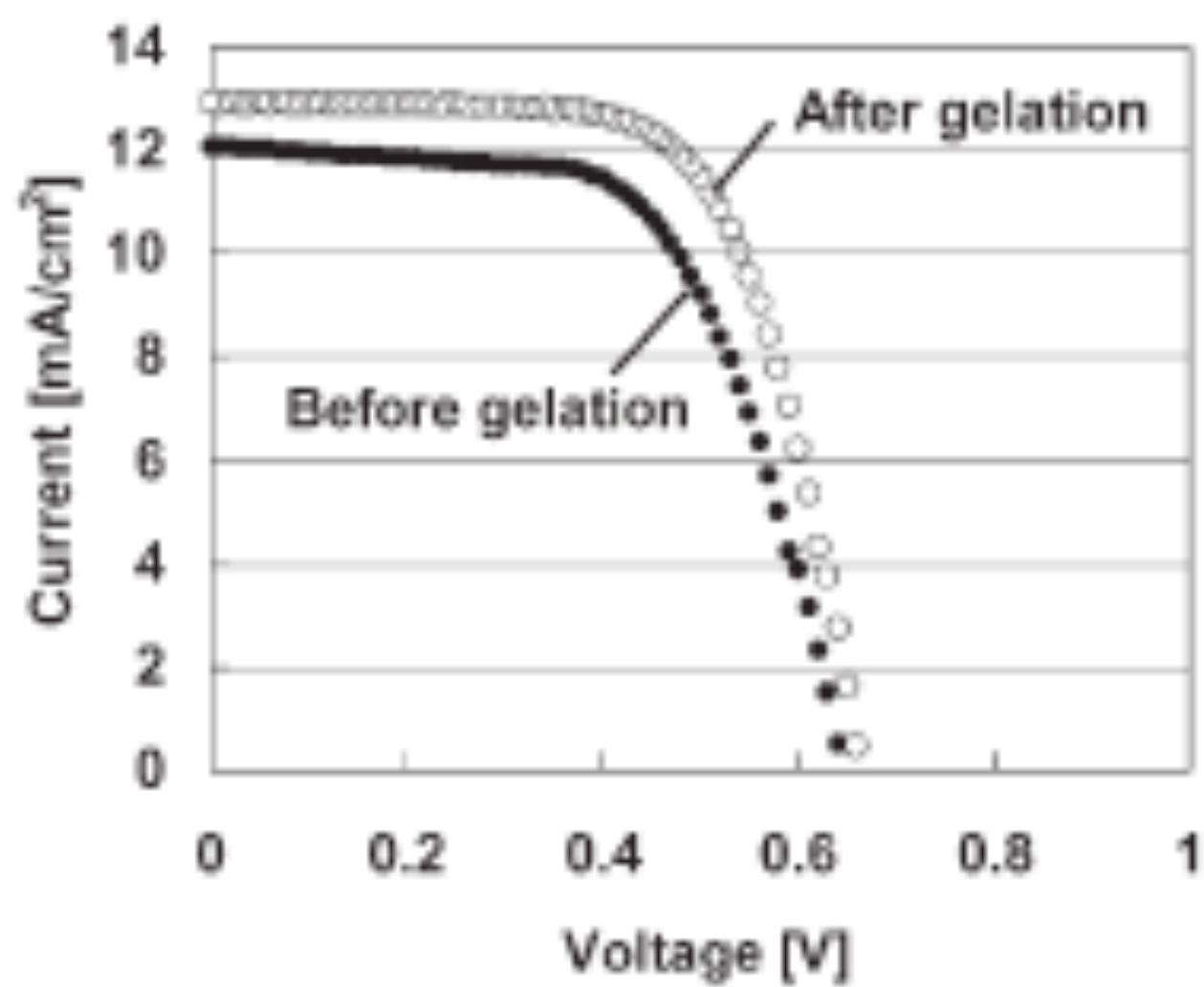
New latent chemically-cross-linked gel electrolyte precursors for quasi-solid dye sensitized solar cells (QDSC) are reported. The gel electrolyte precursors consist of nano-particles and dicarboxylic acids as the latent gelators. The viscosity of the precursor is low at first and does not increase during storage at room temperature. However, when the precursor is baked at 80 °C, it solidifies immediately. Photo-voltaic performance is maintained after solidification.

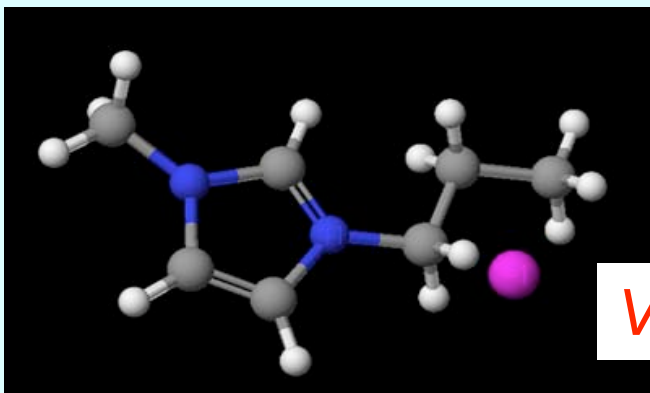
Dye sensitized solar cells (DSC) contain volatile liquid electrolytes.¹ Therefore, achieving solidification is one of the crucial research areas: all-solid DSCs have been reported previously.²⁻¹⁰

Table 1 Summary of gel electrolyte precursors and their compositions

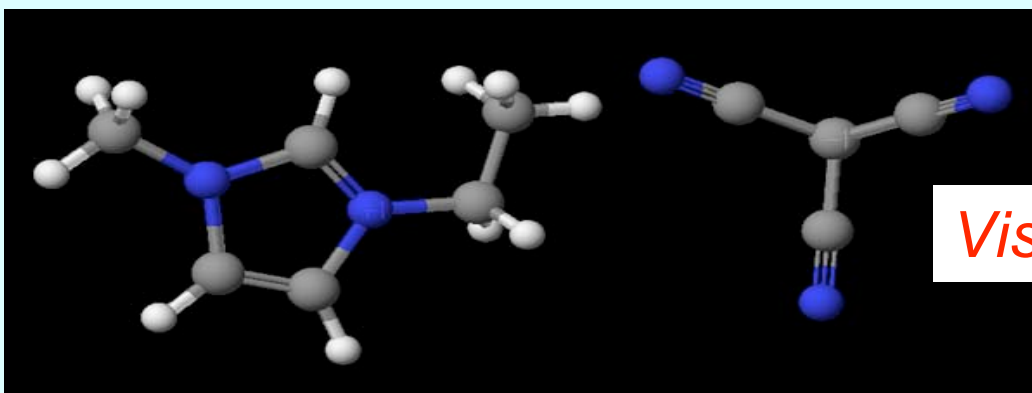
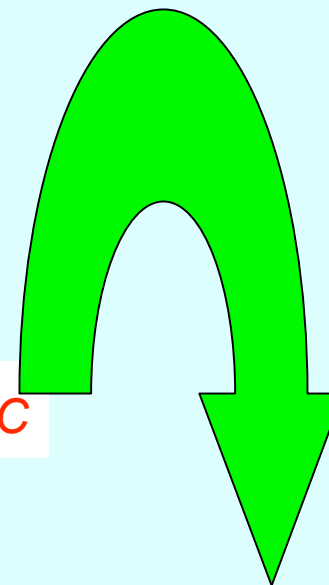
Abbreviation ^a	Electrolyte TS3 ^b	Nano-particle ^c	Dicarboxylic acid	Gelation?
Pregel-C6-300	100	3	3	No ^d
L-Gel-Pre-C12-300	100	3	3	Yes
L-Gel-Pre-C16-300	100	3	3	Yes

^a C6: HOOC(CH₂)₄COOH, C12: HOOC(CH₂)₁₀COOH, C16: HOOC(CH₂)₁₄COOH. ^b Electrolyte (TS3): MePrImI (containing 5% water), I₂ 300 mM, *t*-BuPy 580 mM, LiI 500 mM. ^c AEROSIL 300 (Nippon Aerosil). ^d After a swift reaction, precipitation occurred.



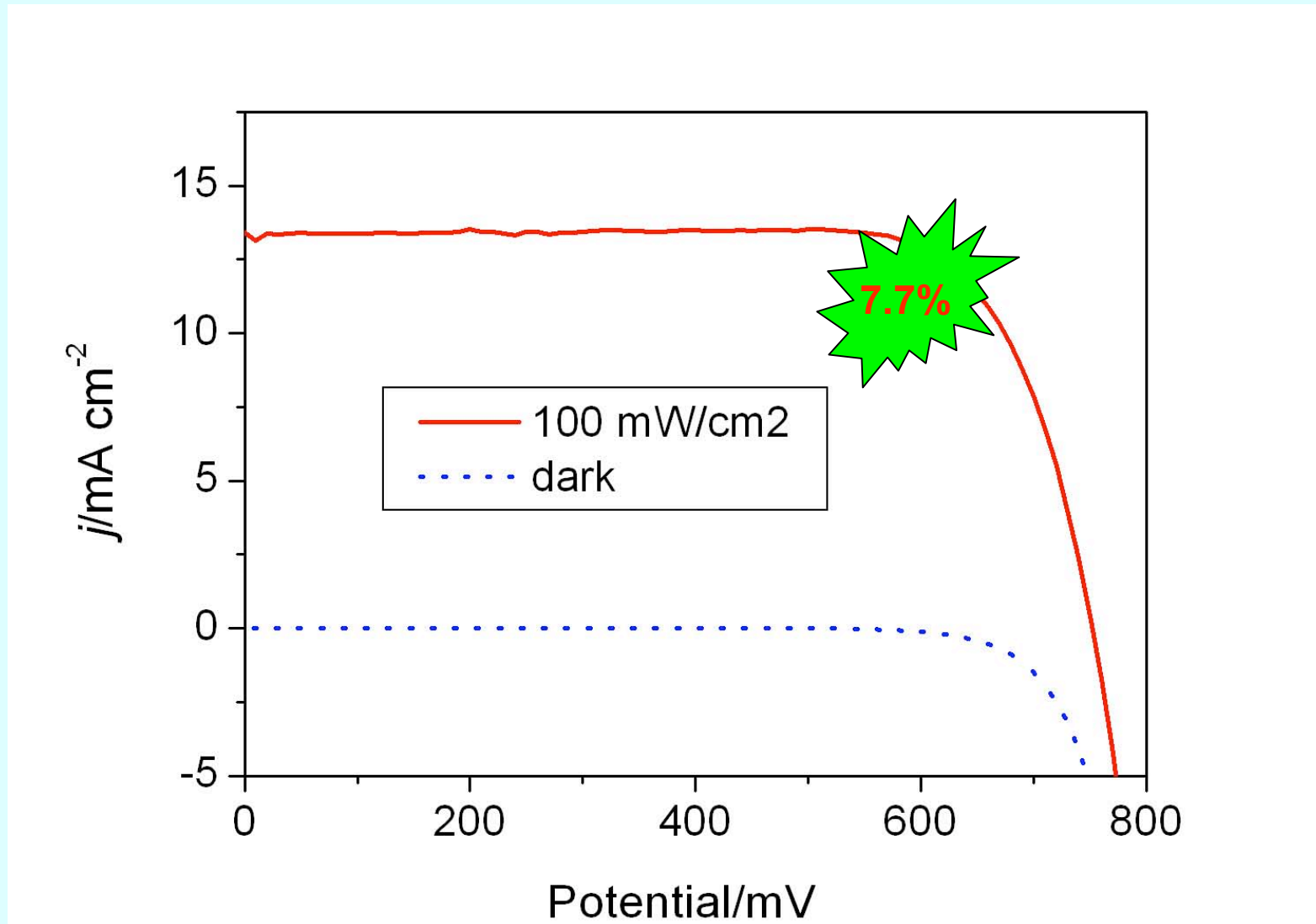


Viscosity: 900 cp at 22 °C



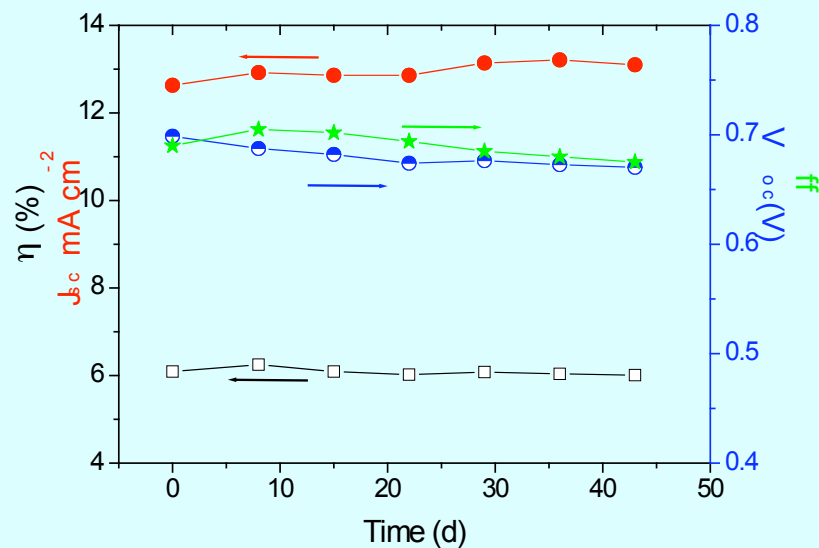
Viscosity: 18 cp at 22 °C

Photovoltaic performance



Wang, P.; Klein, C.; Humphry-Baker, R.; Zakeeruddin, S.M. and Grätzel, M. *J. Am. Chem. Soc.* 2005, 127, 808.

Evolution of device parameters using quasi-solid ionic liquid gel electrolyte under one Sun light soaking at 60 °C



PMII/EMINCS: 65:35 (volume)

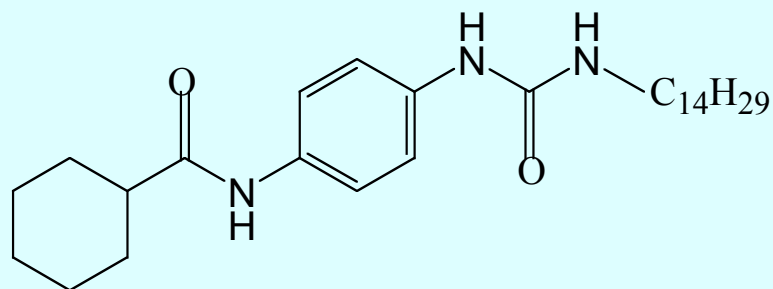
I₂: 0.2 M

NMBI: 0.5 M

GuNCS: 0.1 M

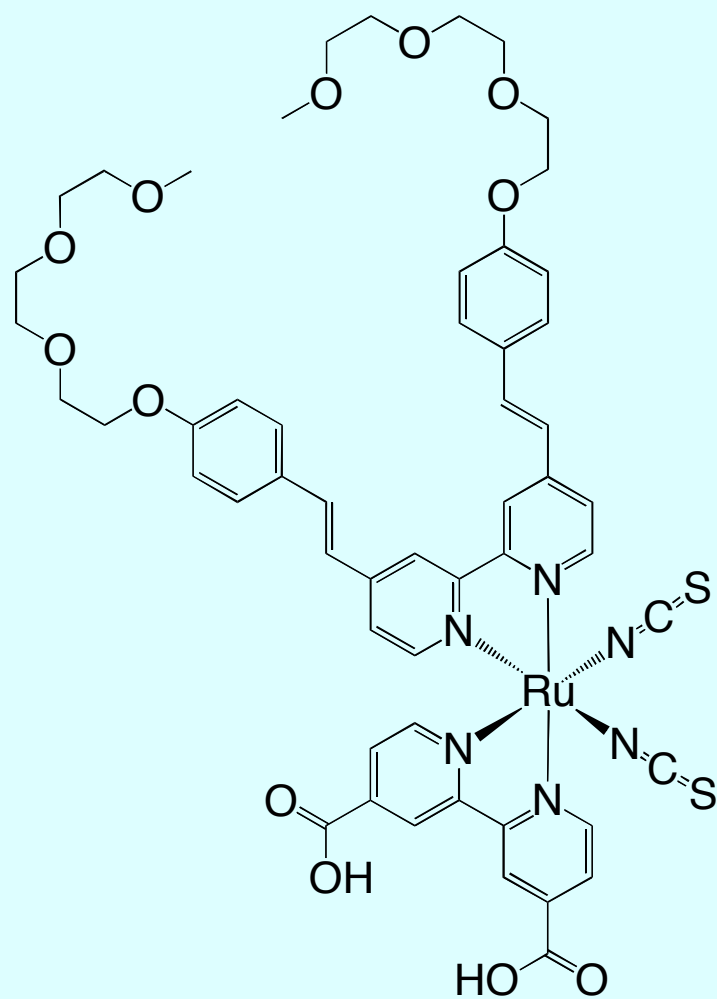
Gelator: 2 wt %

Gelator

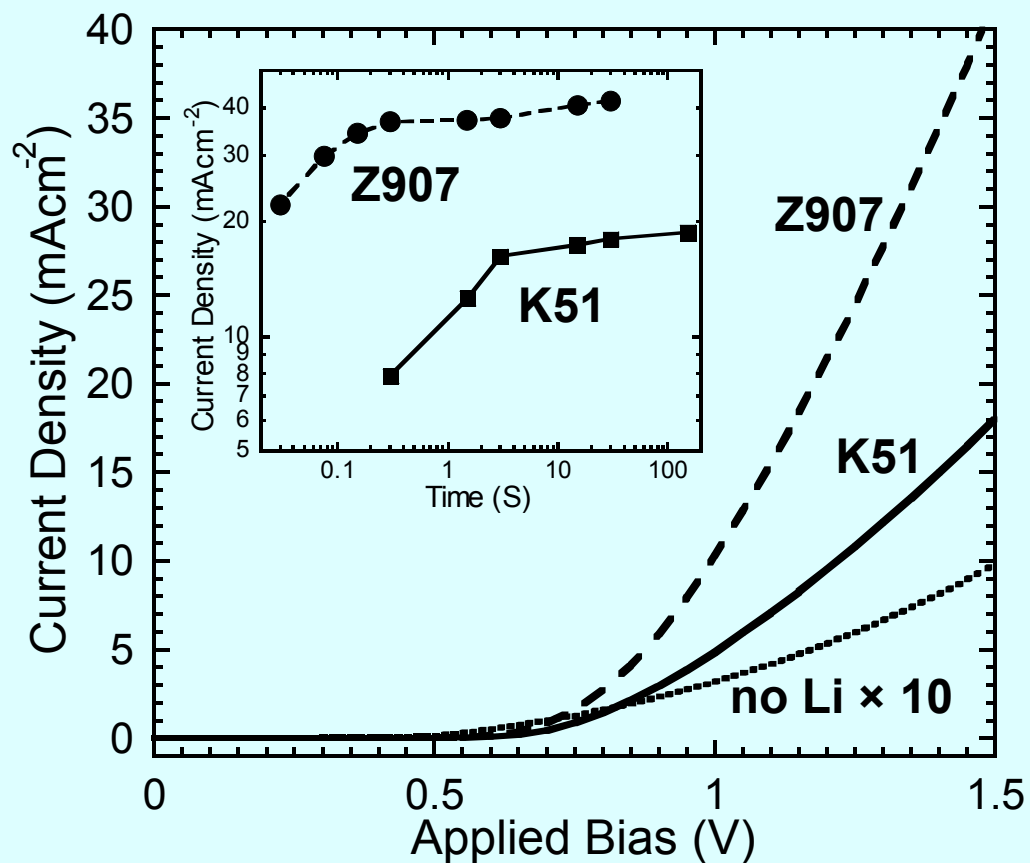


Dye K19+DPA

Ion coordinating sensitizers



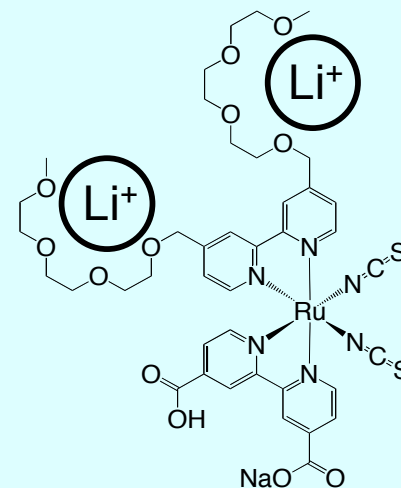
Immobilization of Li Ions



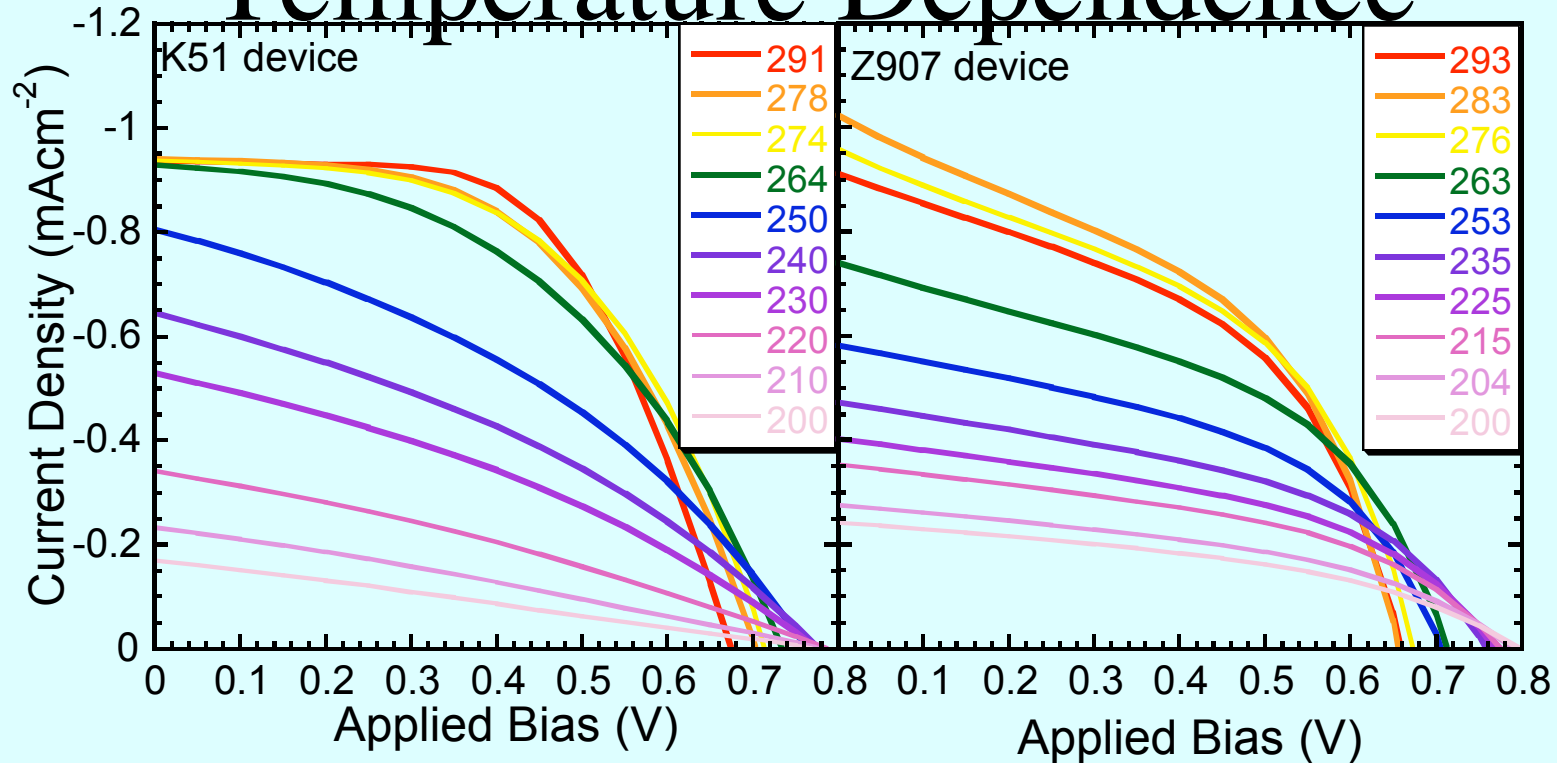
Li hugely increases J density

No longer space-charge limited current.

K51 immobilizes ions on dye backbone.



Temperature Dependence

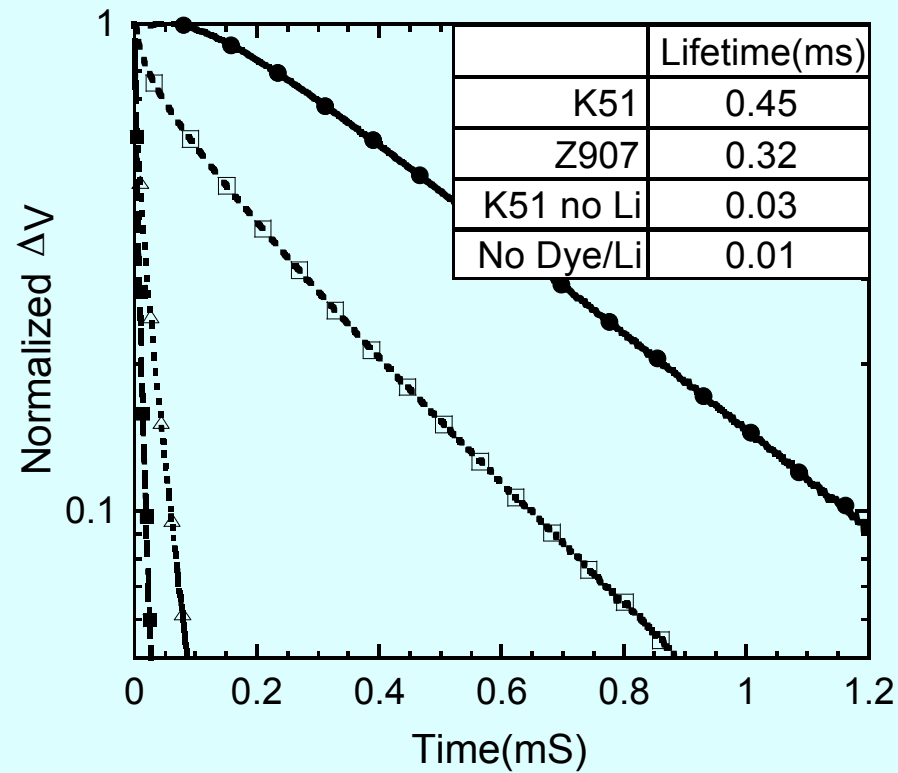


Increased charge mobility with temperature \uparrow current and fill factor

Increased charge recombination \downarrow voltage and current

For K51 \uparrow mobility wins. For Z907 \uparrow charge recombination wins

Transient Voltage Decay's



Research

SHORT COMMUNICATION: ACCELERATED PUBLICATION

Dye Solar Cells Without Electrolyte or Hole-transport Layers: a Feasibility Study of a Concept Based on Direct Regeneration of the Dye by Metallic Conductors

F. O. Lenzmann^{1,*}, B. C. O'Regan¹, J. J. T. Smits², H. P. C. E. Kuipers², P. M. Senneling¹, L. H. Slooff¹ and J. A. M. van Roosmalen¹

¹ECN, Solar Energy, Westenduinweg 3, 1755 LE Petten, The Netherlands

²Shell Global Solutions, Badhuinweg 3, CM 1031 Amsterdam, The Netherlands

Flat structures consisting of dense dye-sensitized TiO₂ films with various materials for dye regeneration (TiO₂/dye/regeneration material) are compared. Au and PEDOT:PSS were tested as metal or metal-like regeneration materials and compared with reference compounds, such as the redox couple I⁻/I₃⁻ in solution and p-type CuSCN. Under the exclusion of TiO₂ bandgap excitation, the short-circuit photocurrent densities for the various structures differ by less than ~30%, suggesting comparable charge separation efficiencies. The good performance of a metallic regeneration material implies, that the frequently assumed requirement of p-type or 'hole conducting' properties for the regeneration material in solid state dye solar cells is questionable. Copyright © 2005 John Wiley & Sons, Ltd.

KEY WORDS: dye-sensitized solar cells; solid state; Au; hole transport layer

1. INTRODUCTION

The development of dye sensitized solar cells¹ over the past years has resulted in improved long-term stability² and increased efficiency. Currently, the highest certified efficiency values are 11.0% for small liquid junction type laboratory cells (0.25 cm²) and 8.2% on larger cell areas (2.36 cm²).^{3,4} Recent progress in device manufacturing indicates that this technology is approaching maturity for market introduction.⁵

* Correspondence to: Frank O. Lenzmann, ECN, Solar Energy, Westenduinweg 3, 1755 LE Petten, The Netherlands.

E-mail: lenzmann@ecn.nl

Contract/grant sponsors: Shell Research Foundation; EZ.

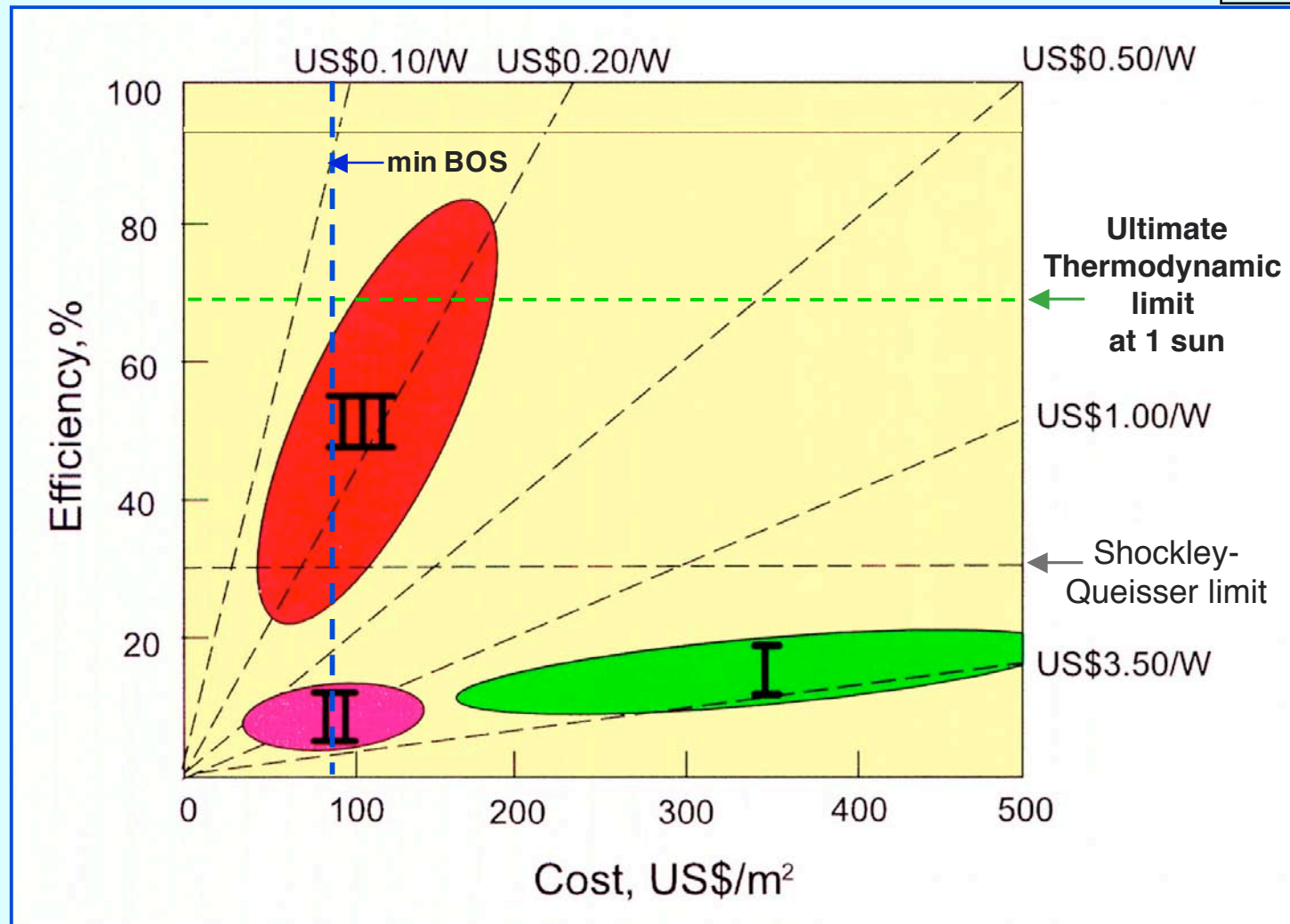
Contract/grant sponsor: European Commission; contract/grant number: SES6-CT-2003-502620.

Advantages vs. Silicon Cells

- **Low cost and ease of production**
- **Performance increases with temperature narrowing the efficiency gap**
- **Bifacial configuration - advantage for diffuse light and albedo**
- **Efficiency less sensitive to angle of incidence**
- **Transparency for power windows**
- **Color can be varied by selection of the dye, invisible PV-cells based on near-IR sensitizers are feasible**
- **Low energy content (for silicon this is 5 GJ/m² !), payback time is only a few months as compared to years for silicon.**
- **Outperforms amorphous Si**

PV Power Costs as Function of Module Efficiency and Cost

From Martin Green



For PV to provide the full level of C-free energy required for electricity and fuel—solar power cost needs to be ~5 cents/kWh (\$1.00 W_p)



®

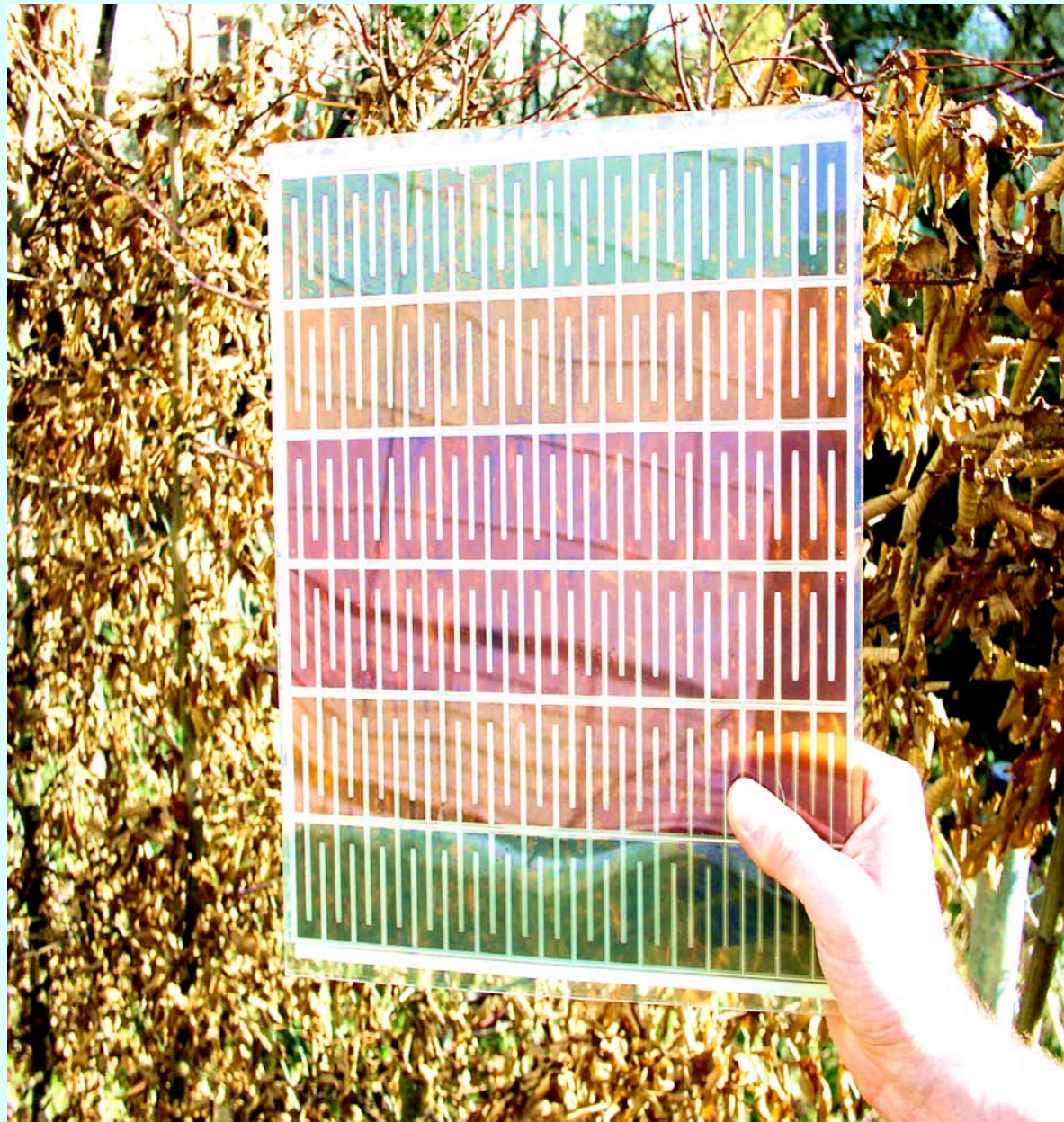
Konarka®





Courtesy of Greatcell Solar

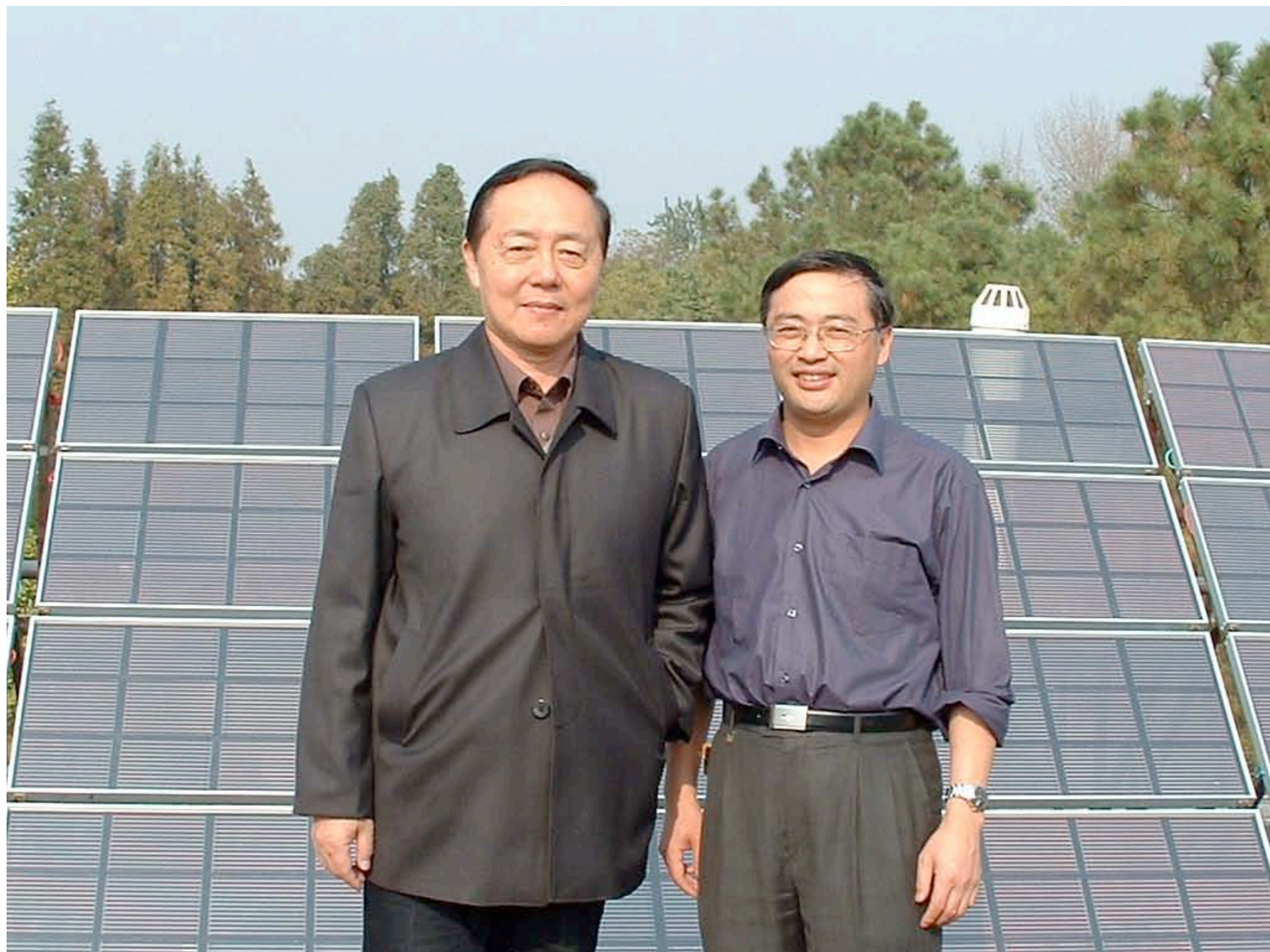
Various colours in a series-connected dye solar cell module



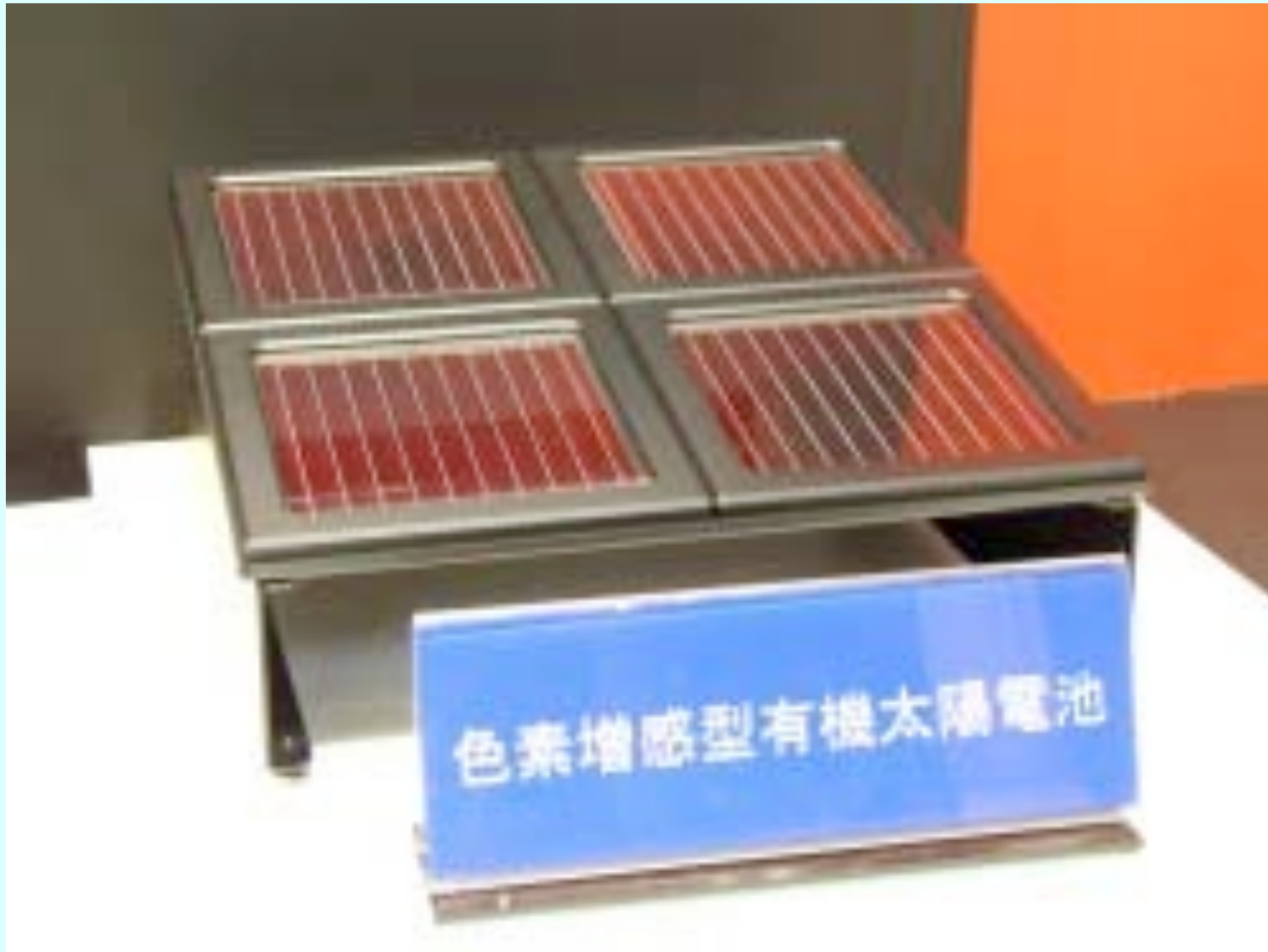
Courtesy Dr. Winfried Hoffman, CEO, RWE, SCHOTT Solar GmbH











Hitachi's new dye sensitized cell achieves 9.3 percent efficiency





温水



特許技術ナノテック
Adaptive Refrigerator
打ち水の原理で冷やす
クリーンなエアコン。

This type of clean air conditioner uses the principle of sprinkling water on a surface to cool it (evaporative). ナノテックが組み出した超微細な、水分子を吸着する活性炭の多孔質材料は水を吸い取り、水分子を蒸発させます。

nanotech



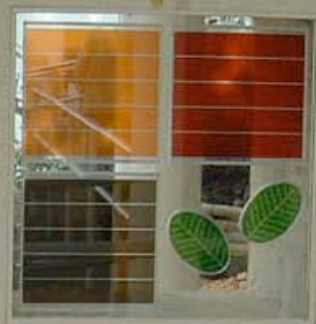
「エナジーガーデン」
様々なエネルギーを上手に使うことによって、
CO₂の発生を抑えながら快適に暮らすことが可能です。

“Energy Garden”

By better using different types of energy, we can lead comfortable lives while reducing CO₂ emission.

室内エアコン

送風用



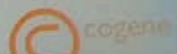
家庭内の家電

New Energy
For a b

FUEL CELL COGENERATION SYSTEM

家庭型燃料電池
Stationary Fuel Cells
わが家では、電気もお湯も
水素と酸素からつくる。

In the house, both electricity and hot water are produced using hydrogen and oxygen.
都市ガスからつくった水素と、空気の酸素を使って発電します。
その発電でつくられるお湯もわかします。



色変換型太陽電池
Dye-sensitized Solar Cells
大自然の植物が教えてくれた。
新しい太陽電池。

A new type of solar cell inspired by Mother Nature's plants.
植物が光合成をするときに、色素で光エネルギーを分解することによって、そのエネルギーを電気に変換しました。





The Toyota Dream House

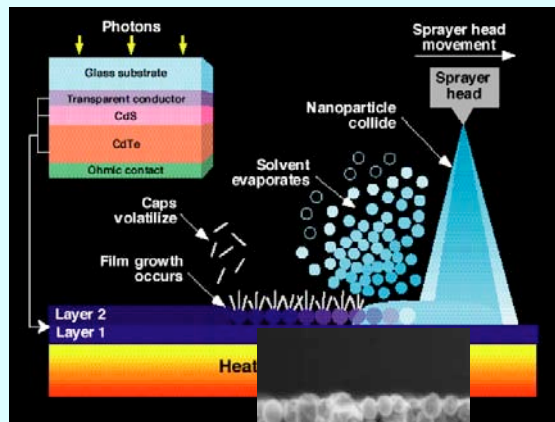
**DSC
made by
AISIN -SEIKI**

http://www.toyota.co.jp/jp/news/04/Dec/nt04_1204.html

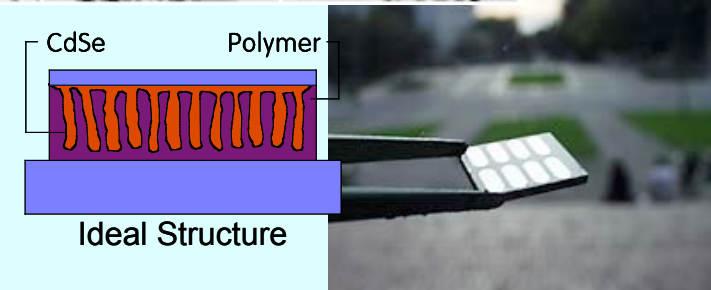
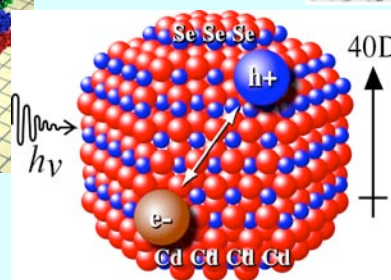
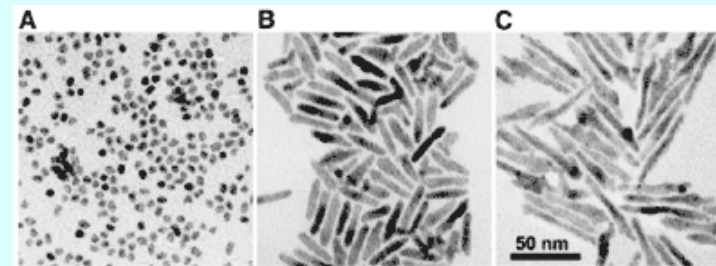
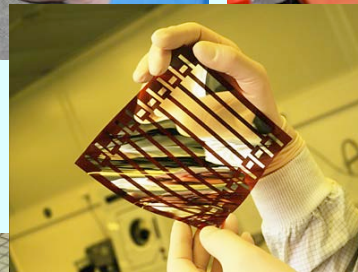
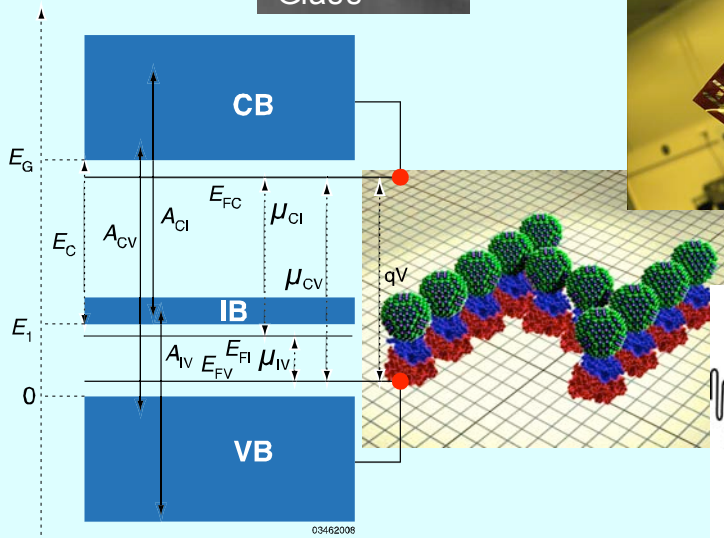
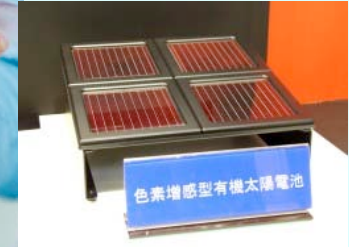
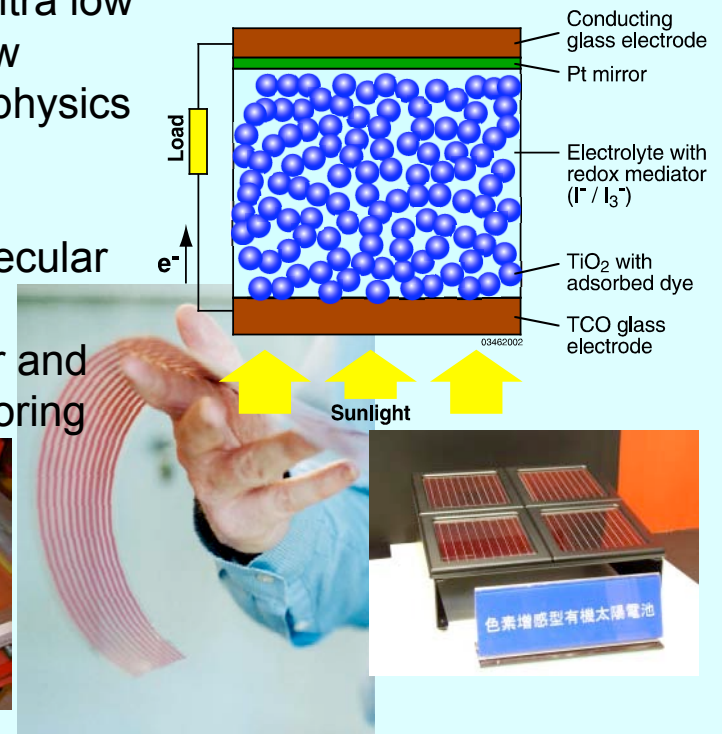
Future Generation PV Technologies

From T. Surek

- Opportunities for innovation– ultra low cost, ultra high efficiencies, new materials, new chemistry, new physics
- Photoelectrochemistry
- Excitonic cells (dye cells, semiconducting polymers, molecular semiconductors)
- Quantum dot cells; hot-carrier and impurity band cells; photon tailoring



Nanoparticulate
CIGS
precursor
materials
Mo
Glass



Can laptops run on spinach ?



Spinach photosynthetic power
can create electricity.

Integration of Photosynthetic Protein Molecular Complexes in Solid-State Electronic Devices

Rupa Das, Patrick J. Kiley, Michael Segal, Julie Norville, A. Amy Yu, Leyu Wang, Scott A. Trammell, L. Evan Reddick, Rajay Kumar, Francesco Stellacci, Nikolai Lebedev, Joel Schnur, Barry D. Bruce, Shuguang Zhang, and Marc Baldo

Nanoletters 2004, vol 4, pp 1079 - 1083;