

NANOBIOSENSORS AS EFFICIENT AND SUSTAINABLE TOOLS FOR DEMOCRATIZING OUR CURRENT ENVIRONMENT MONITORING SYSTEM

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NanoAlb, Tirana, Albania

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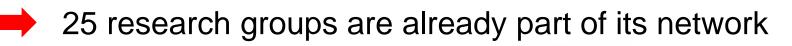


What is NanoAlb?

- Albanian NanoScience and Nanotechnology Unit
- Created in 2019 next to and with the support by the Academy of Sciences of Albania.
- It is a virtual center that coordinates the activities in the area of nanoscience and nanotechnology in Albanian universities in Albania, Kosova, North Macedonia and Montenegro.







EPOKA



Nanotechnology importance for Albanian and Western Balkans economy

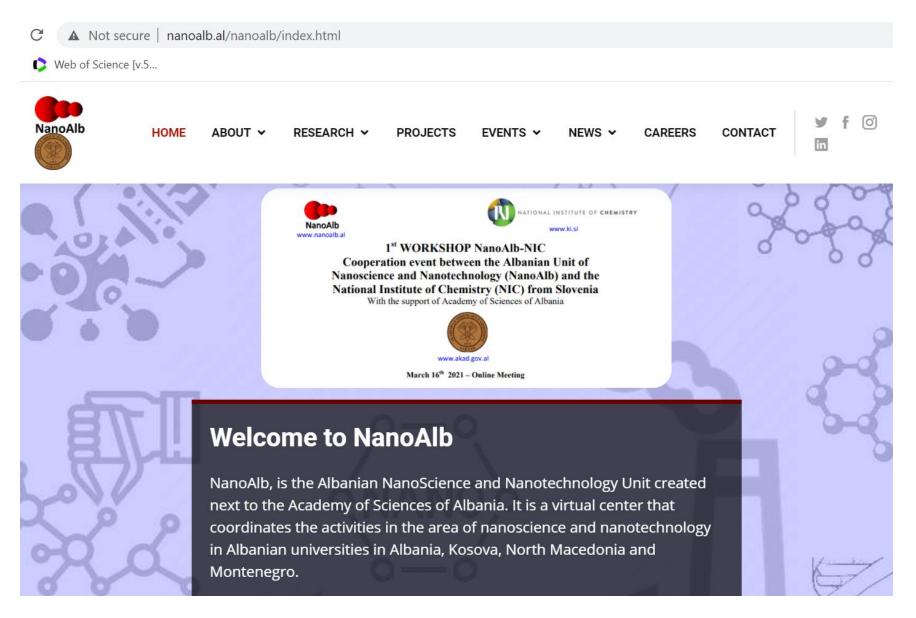
- Applications in HEALTH, AGRICULTURE, ENVIRONMENT, SAFETY AND SECURITY and other industries
- It is a transversal field of research (englobing from physics, chemistry, biochemistry, material science, electronics, medicine, agriculture etc.)
- Societal impact
- Opening a new model of science in Albania and region (highly competitive research centres)
- Attracting & retaining talent
- Attracting investment from international high tech industries
- Building a new vision:

science /high-tech friendly country & region

- Industrial impact.
- Serving as hub of spinoff companies and other links with various industries in the region.

Opening of new working places for professionals

More info on NanoAlb at the website www.nanoalb.al



Catalan Institute of Nanoscience and Nanotechnology (ICN2)

Trustees:

Generalitat de Catalunya

SCSIC UAB

Center of:

BIST Barcelona Institute of Science and Technolog

Member of:

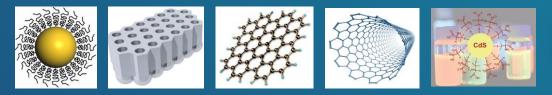
SOMM EXCELLENCE ALLIANCE

ICN2⁹ excelencia severo ochoa

NANOBIOELECTRONICS AND BIOSENSORS

The Group

Focuses on **discovering** and on the **technological development** of cuttingedge nanotechnology and nanomaterials towards **simple and costeffective nanobiosensing platforms.** We do this controlling their architectures at both **nano** and **macro** scales with the objective to link together these two worlds.



Application areas:

- ✓ Diagnostics
- Environmental Monitoring
- Food quality
- ✓ Safety / security
- Other industrial applications

Technology transfer:

paperdr()pdx



OUR RESEARCH

Nanomaterials

Graphene and 2D materials, Metal nanoparticles, quantum dots

Paper-based

Lateral flow assay, hybrid electrochemical/LFA, optical redout



Printing technologies

Inkjet printing, screen printing, graphene transfer electrodes, FET-based biosensors, electrochemical readout

Fully integrated PoC devices Smartphone based, wearables, wireless readout

OUR CHALLENGES

1 Can we make diagnostic devices completely non-invasive?



2 Can we ensure continuous / real time (bio)monitoring?

③ Which partners & expertise's we need to develop devices for real applications that generate (bio)data?



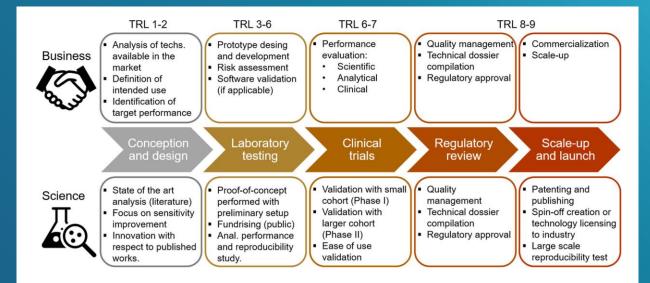
How to make cheap/low cost devices that can be disposable?

6 How to correlate (sensor)data to get insights about the body?

How COVID19 changed the scenario

The development of new diagnostic devices is a process characterized by several bottlenecks. Most of them related to sub-optimal interactions between the actors involved, i.e.:

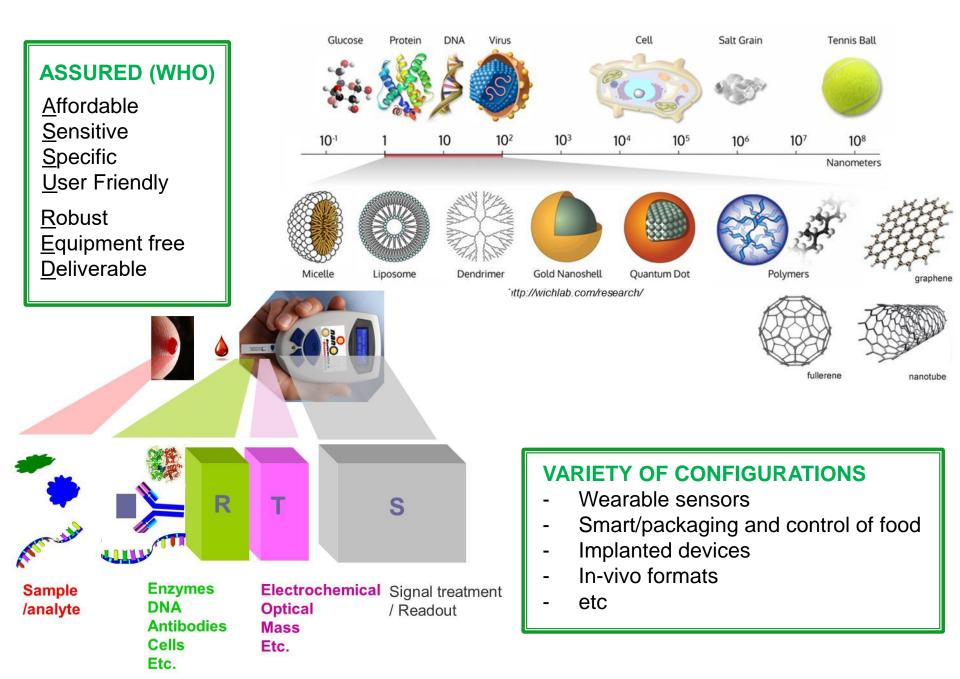
- The scientists
- The clinics
- The regulatory bodies
- The companies



What can we do to change the status quo?

Rosati, Merkoçi et al. 2021 Nanodiagnostics to Face SARS-CoV-2 and Future Pandemics: From an Idea to the Market and Beyond ACS Nano 2021, 15, 11, 17137–17149

PLENTY OF POSSIBILITIES FOR NANOBIOSENSORS



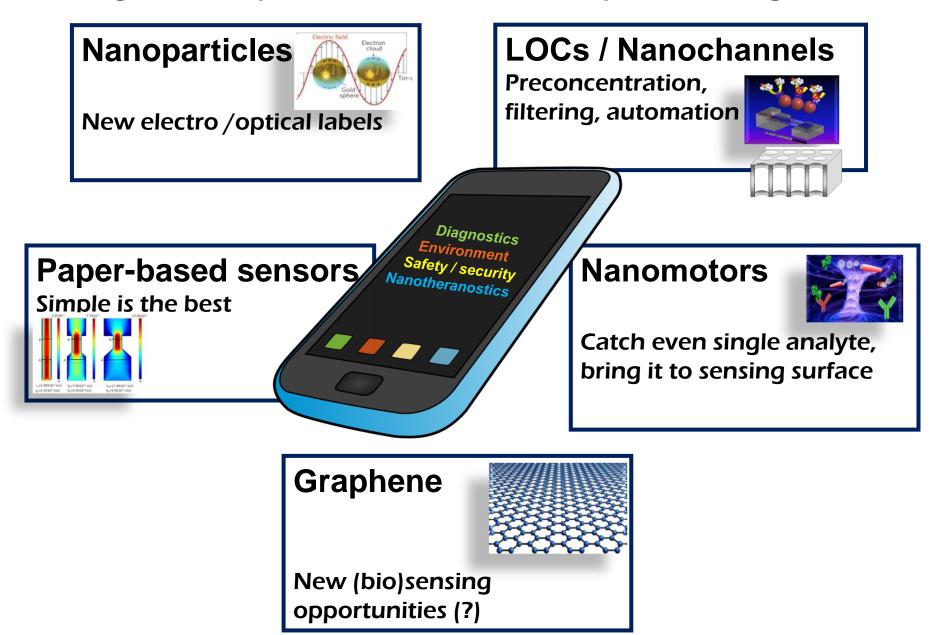
NANOBIOELECTRONICS AND BIOSENSORS



Institut Català de Nanociència NAN i Nanotecnologia **Objectives** Bioelectronics & Biosensors Group **Diagnostics** N **Environmental** A monitoring N D **NANOBIOELECTRONICS &** 0 Ε **BIOSENSORS**'s research aims M to integrate nanotechnology Other methods, tools and materials into industrial low cost, user friendly and C applications efficient (bio)sensors with Ε Ε interest for diagnostics, safety R S /security and other fields Food Α quality Safety / security S

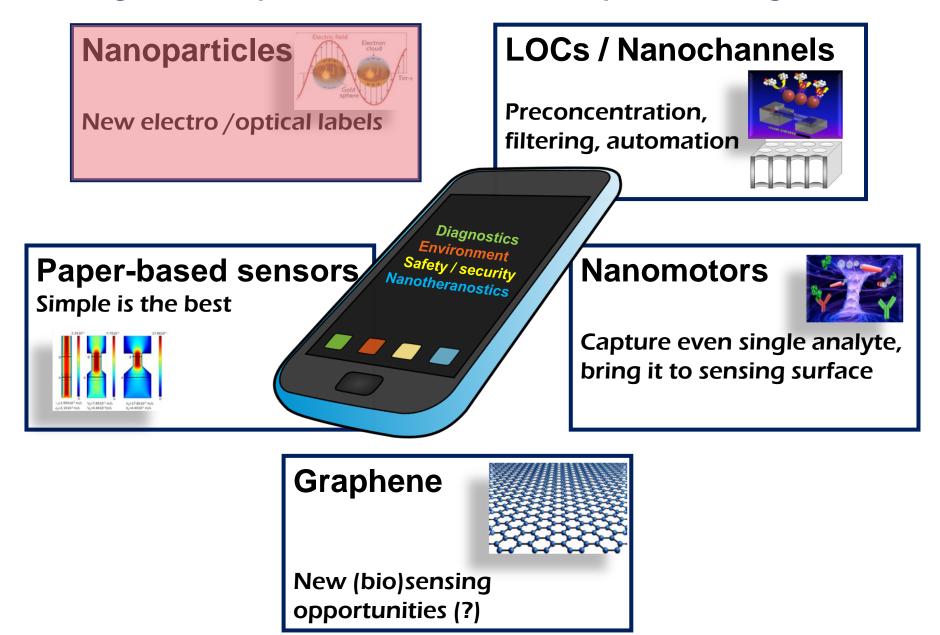
OUR MOTIVATION

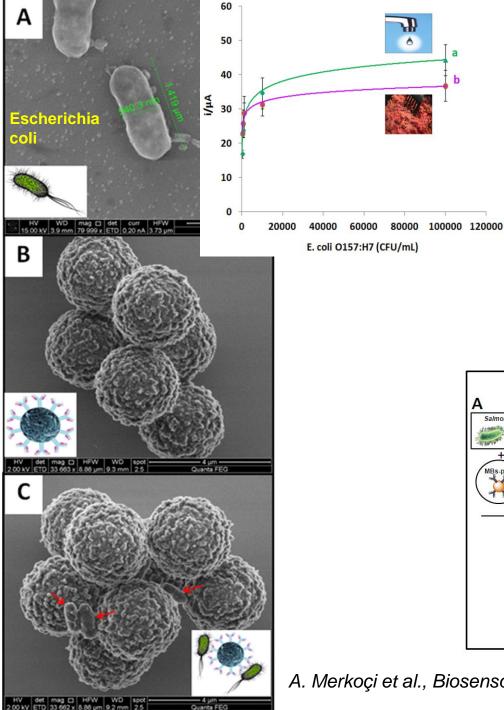
To design new simple nanobiosensors and improve existing ones



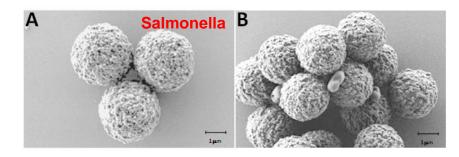
OUR MOTIVATION

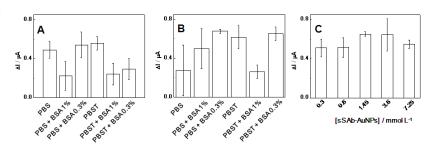
To design new simple nanobiosensors and improve existing ones

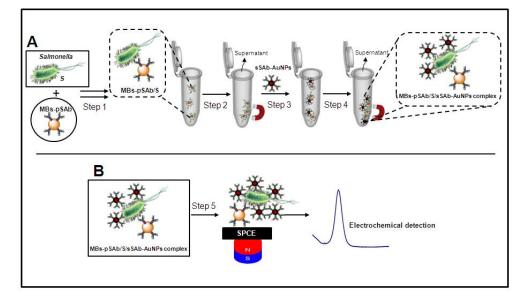




Au-NP-based detection of bacteria





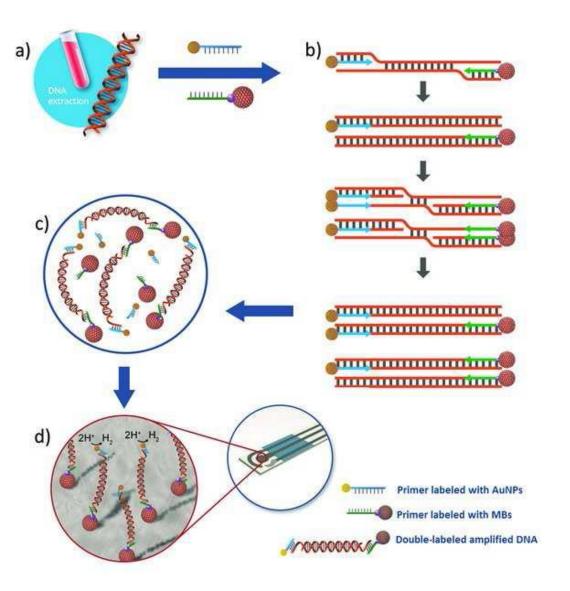


A. Merkoçi et al., Biosensors and Bioelectronics, 2013, 40, 121–126

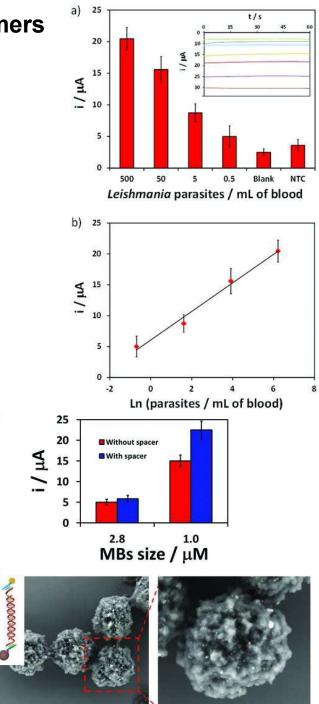
a

b

Magnetic Bead/Gold Nanoparticle Double-Labeled Primers for Electrochemical Detection of Isothermal Amplified *Leishmania* DNA



Escosura, Pires, Merkoçi et al., Small 2016, 12, 205–213



a)

b)



Cite This: Anal. Chem. 2019, 91, 4790-4796



In Situ Plant Virus Nucleic Acid Isothermal Amplification Detection on Gold Nanoparticle-Modified Electrodes

Mohga Khater,^{†,‡} Alfredo de la Escosura-Muñiz,^{†©} Laura Altet,[§] and Arben Merkoçi*^{,†,∥}⊙



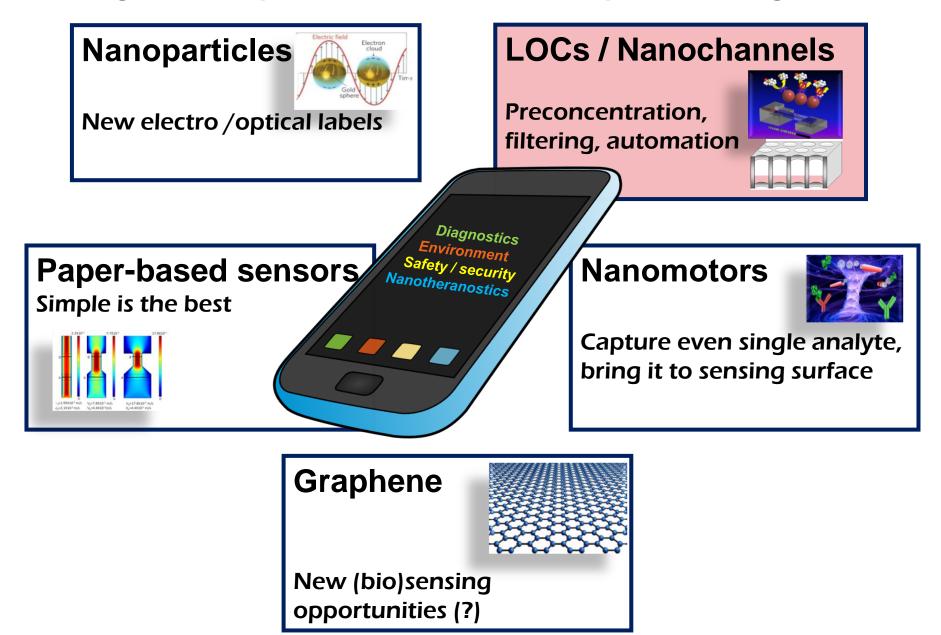
Detection of citrus tristeza virus (CTV)

Z" (D) Z' (Ω) Z' (Ω) TITT 1111 AuNP-modified SPCE CTV-p20 gene Recombinase Polymerase Amplified dsDNA RPA primers SSB

The in situ-amplified CTV target was investigated by EIS in a $Fe(CN_6)^{4-}$ /Fe(CN₆)³⁻ red–ox system, being able to quantitatively detect <u>1000 fg µL⁻¹</u> of nucleic acid. High selectivity against nonspecific gene sequences characteristic of potential interfering species such as *Citrus psorosis* virus (CPsV) and *Citrus caxicia* viroid (CCaV) was demonstrated. Good reproducibility (<u>RSD of 8%</u>) and long-term stability (up to 3 weeks)

OUR MOTIVATION

To design new simple nanobiosensors and improve existing ones



Chip fabrication and electrode integration

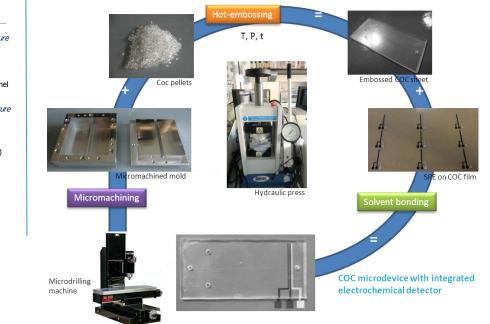
PDMS Chips

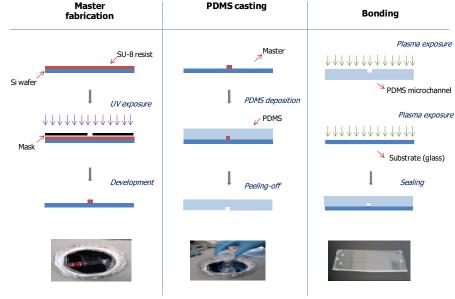
softlithography process

COC Chips (Collaboration. J.L.Viovy)

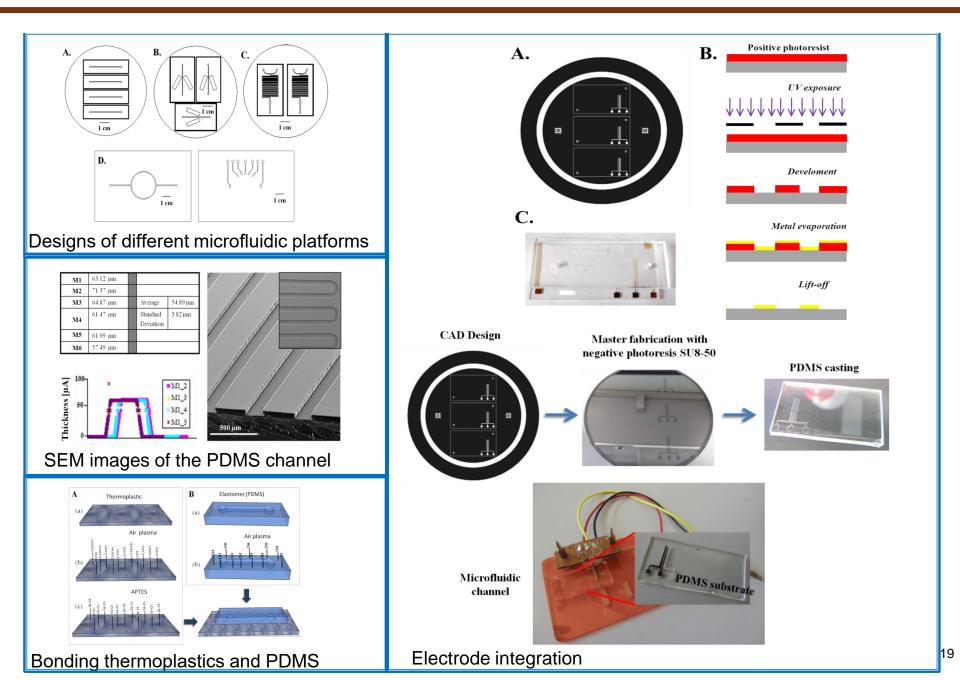


Simple and low cost fabrication techniques



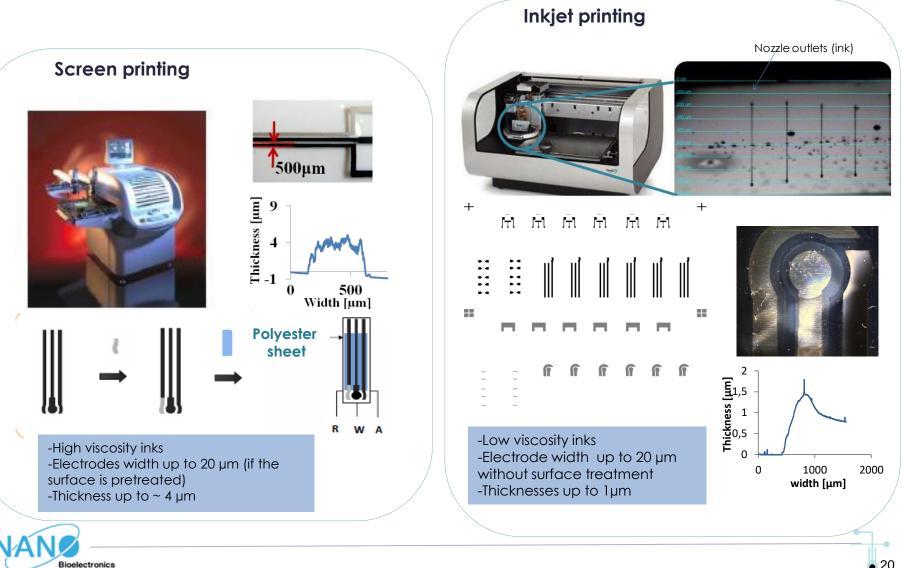


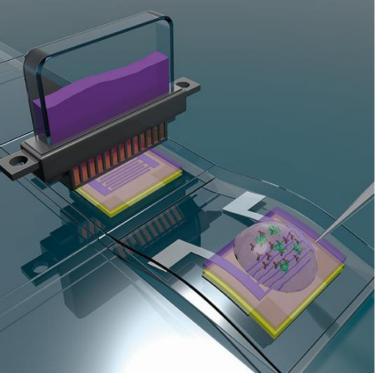
Chip fabrication and electrode integration



Electrode fabrication

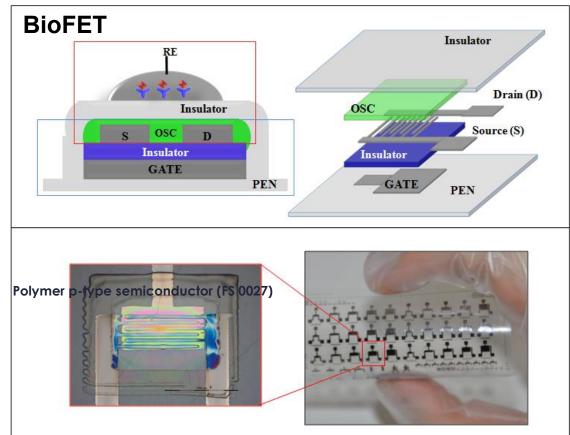
& Biosensors Group





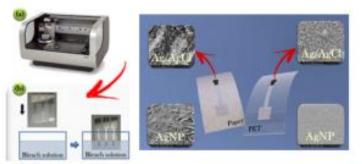
Inkjet-printed sensing platforms using nanomaterial-based inks and other materials

No need for clean room at all!



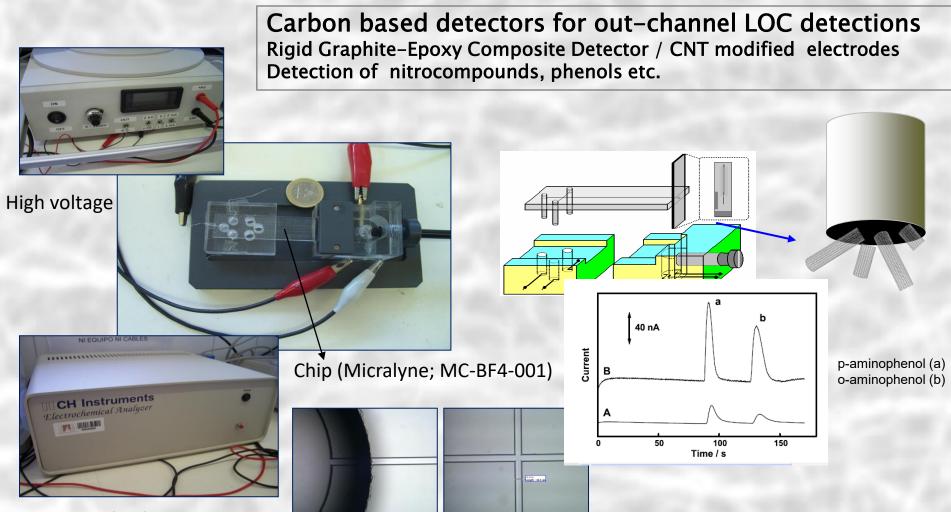
Merkoçi et al. Advanced Functional Materials, 20, 6291–6302. 2014

AgNP-ink jet printed reference electrode in paper or plastic



Merkoçi et al. Anal. Chem. 86, 10531-10534. 2014

CNT / LOC and capillary electrophoresis-based analysis

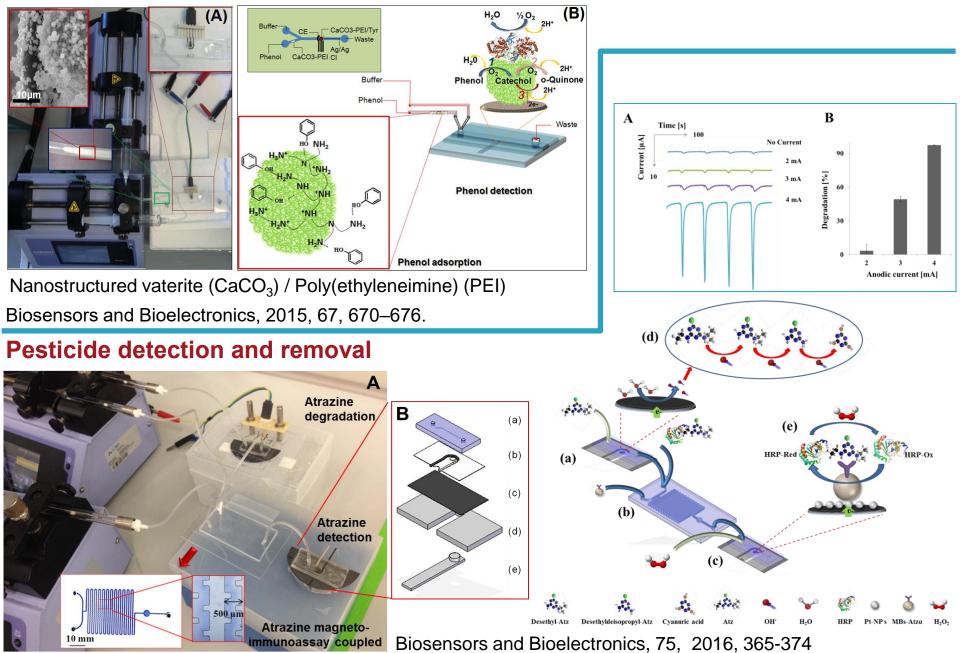


Measuring instrument

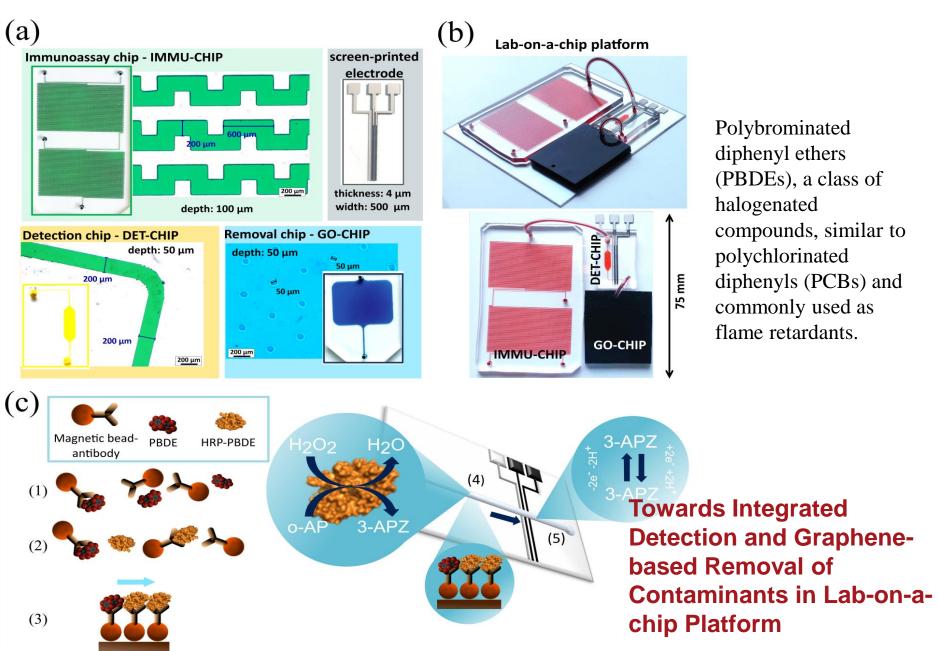
Microchimica Acta, 152, 261–265, 2006 Electrophoresis 27, 5068–5072, 2006 Electroanalysis 18, 207 – 210, 2006 Electrophoresis 28, 1274–1280, 2007

BIOSENSOREMOVAL NanoTechnologies

Phenol detection and removal

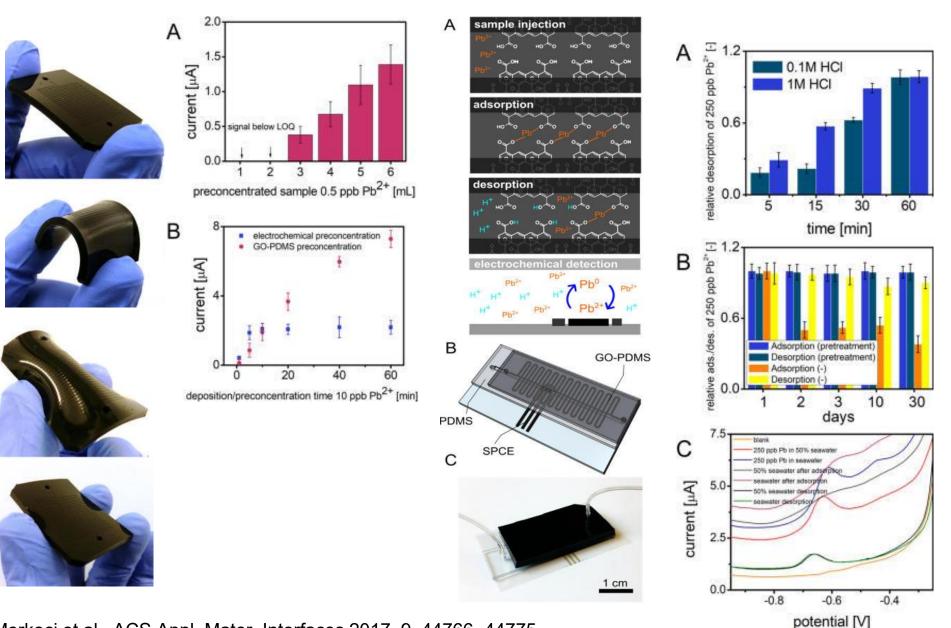


BIOSENSOREMOVAL NanoTechnologies



Merkoçi et al., Nano Research, 2017, Vol 10 (7), pp 2296-2310

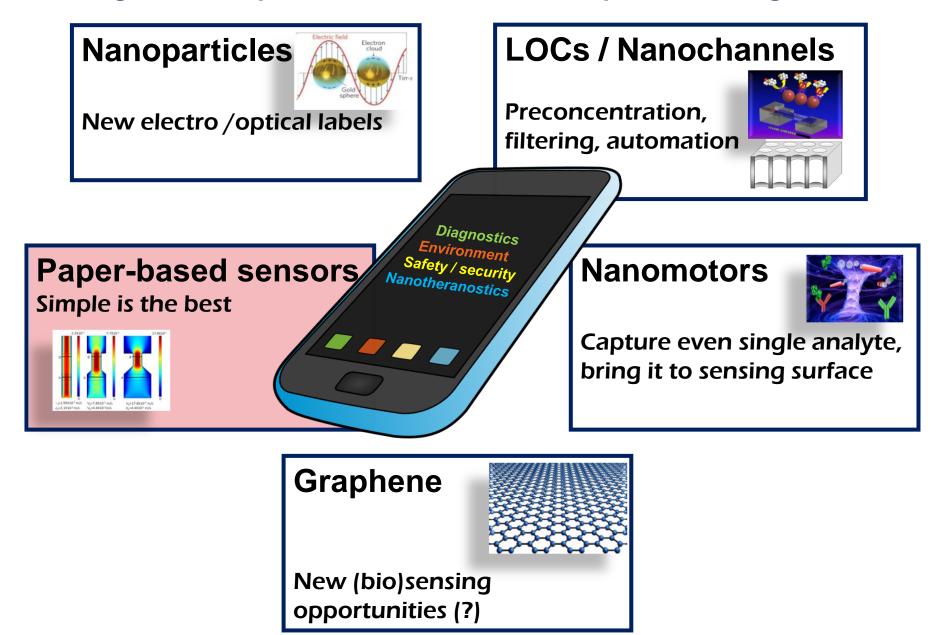
Graphene Oxide–Poly(dimethylsiloxane)-Based Lab-on-a-Chip Platform for Heavy-Metals Preconcentration and Electrochemical Detection



Merkoçi et al., ACS Appl. Mater. Interfaces 2017, 9, 44766-44775

OUR MOTIVATION

To design new simple nanobiosensors and improve existing ones



Paper-based biosensors



Why to move biosensors to paper format?

Paper...

... is formed by cellulose.

- · Low-cost and aboundant material.
- Easy to manufacture
- Recyclable & biosustainable.

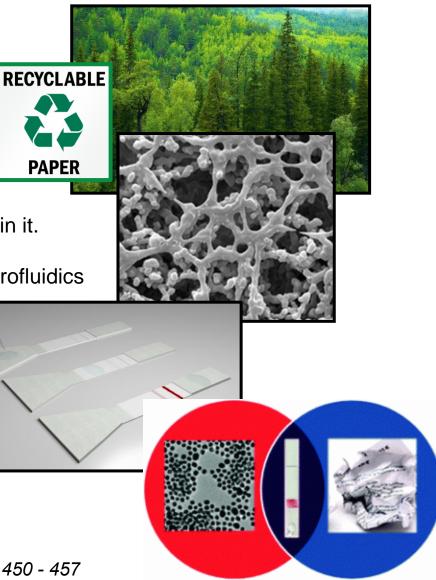
...has a porous matrix.

- Several reactions can be carried out within it.
- The porosity can be modified.
- Capillary forces creates autonomous microfluidics making "zero energy" device!
- ... is easily tunable.
 - Its microfluidics by porosity.
 - Its architecture.

... is compatible with nanomaterials

- Printing of nanomaterials
- Easy nanoplasmonics

Claudio Parolo, Arben Merkoçi. Chem. Soc. Rev., 2013, 42, 450 - 457



Paper based biosensors





- Lateral Flow strips
- Microfluidic devices









http://www.cliawaived.com



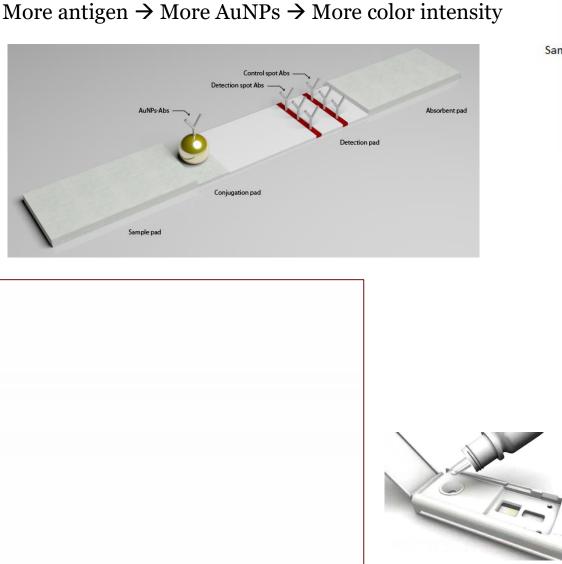
www.dfa.org

Type of paper-based biosensor	Possible detection methods	Advantages	Disadvantages
Dipstick	• Optical	 Easy design Fast optimization	Just one stepOnly optical detectionMostly no quantification
LFA	 Optical Electrochemical	VersatileFlowElectrochemical detectionPossible quantification	 Long optimization times Long fabrication Sample volume (around 100 μL)
μPAD	 Optical Electrochemical Chemiluminescence MEMS 	 Versatile Flow Different detection methods Quantification Small sample volume (less than 10 μL) Massive production 	• Long optimization times

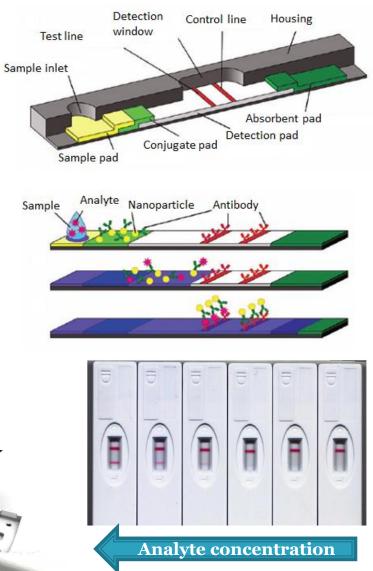
Parolo, Merkoçi Chem. Soc. Rev., 2013, 42, 450

NP-based lateral flow immunoassay



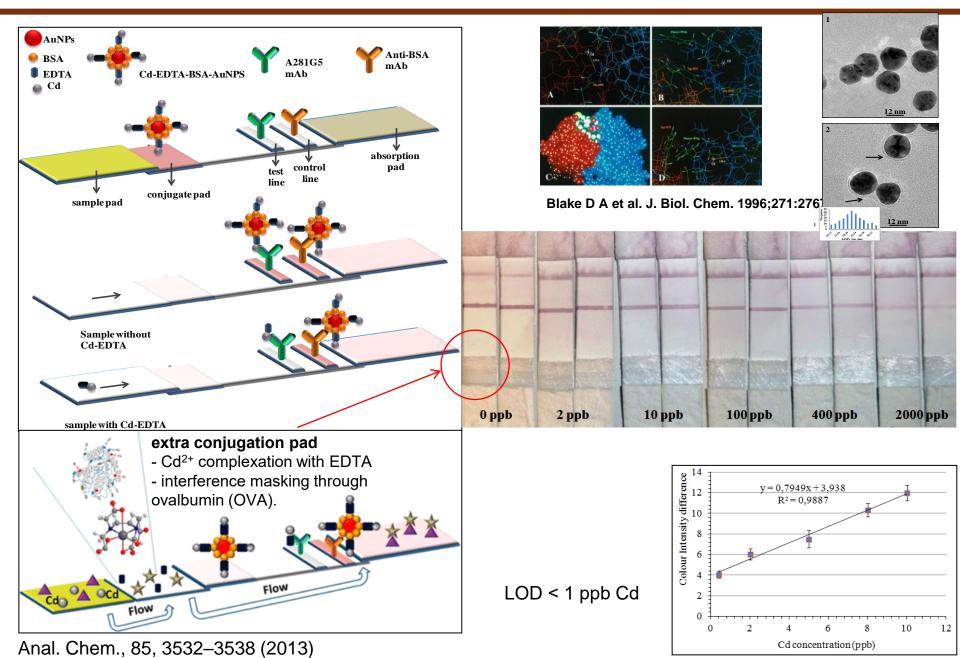


http://www.bbiinternational.com/

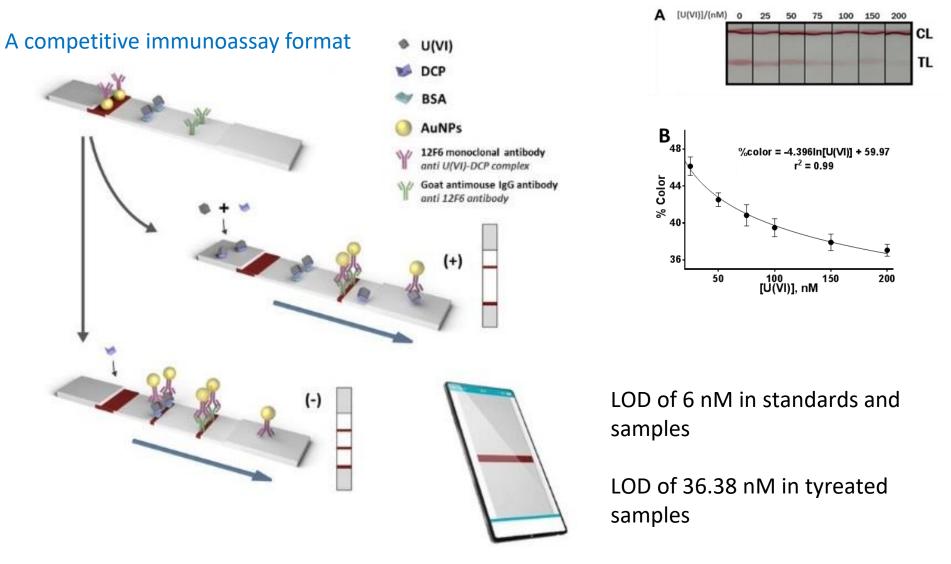


Chem. Soc. Rev. (2010) 39, 1153

LFIA (Cadmium determination in drinking water)

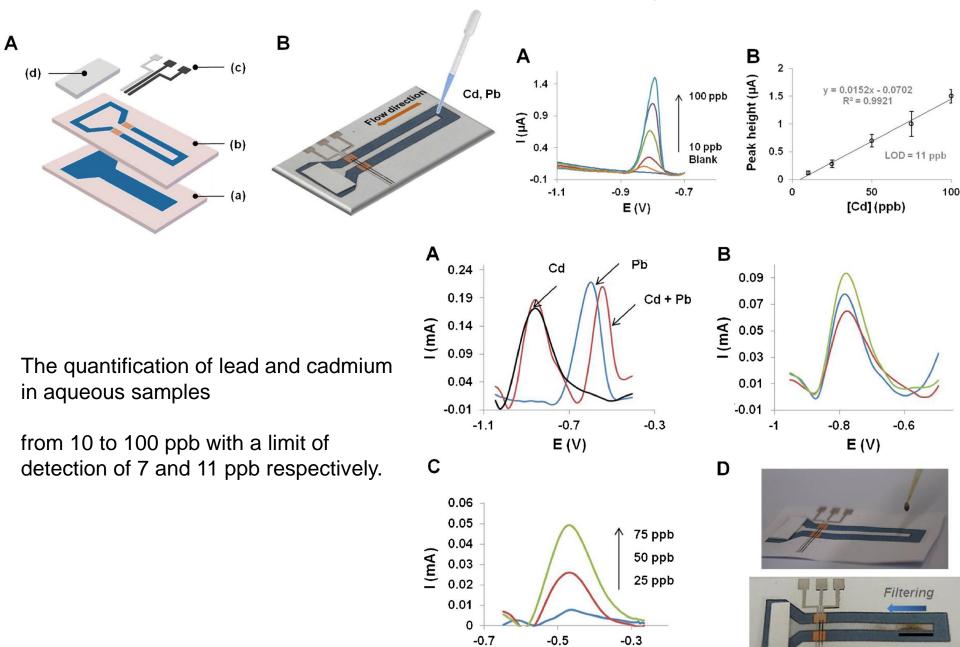


Uranium (VI) detection in groundwater using a gold nanoparticle/paper-based lateral flow device



Merkoçi et al., Scientific Reports volume 8, Article number: 16157 (2018)

Electrochemical lab-on-paper for heavy metal detection



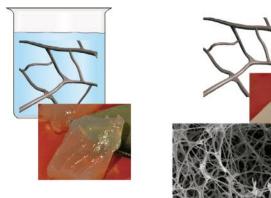
E(V)

~ 1 cm

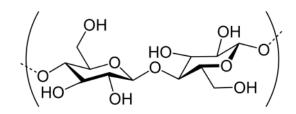
Merkoçi et al., Anal Bioanal Chem (2015) 407:8445–8449

Bacterial Cellulose Nanopaper

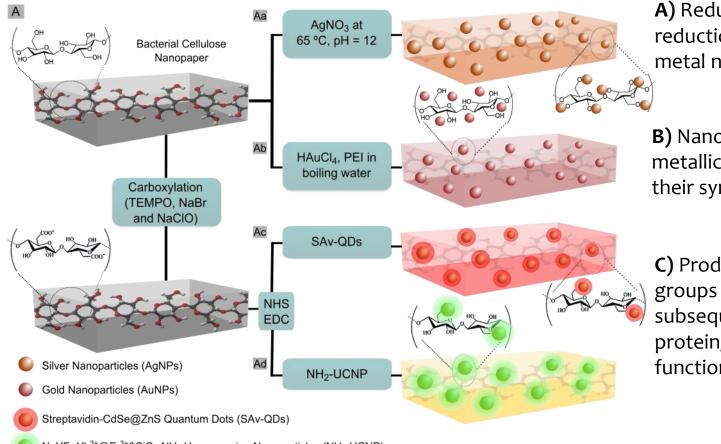
- Multifunctional Biomaterial
- Hydrophilicity
- High porosity
- Broad chemical-modification capabilities
- High surface area



Nat Nanotech 2010, 5:584



Nanopaper as an Optical Sensing Platform Pathways to obtain Plasmonic / Photoluminescent Nanopaper



A) Reducing agent for chemical reduction of noble metal ions to metal nanoparticles

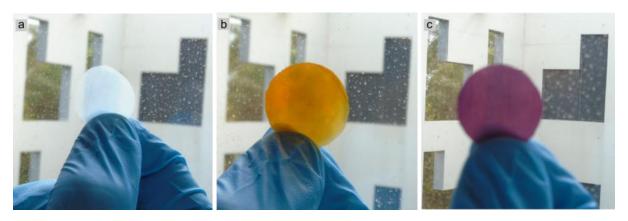
B) Nano-network to embed metallic nanoparticles during their synthesis

C) Producing carboxylic groups on the cellulose, for subsequent coupling with protein/aminofunctionalized nanoparticles

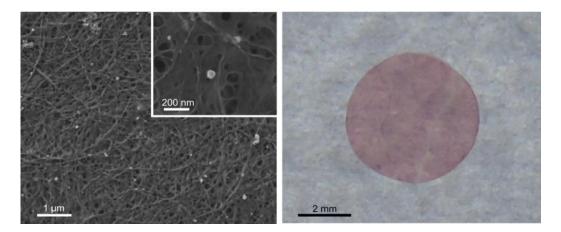
NaYF₄:Yb³⁺@Er³⁺&SiO₂-NH₂ Upconversion Nanoparticles (NH₂-UCNP)

<u>E. Morales-Narváez</u>, <u>H. Golmohammadi</u>, T. Naghdi, H. Yousefi, U. Kostiv, D. Horak, N. Pourreza, A. Merkoçi, ACS Nano **2015**, 9:7296

Nanopaper as an Optical Sensing Platform Plasmonic Nanopaper (appearance)



a. Bare BC. b. AgNP-BC. c. AuNP-BC.



E. Morales-Narváez, H. Golmohammadi, T. Naghdi, H. Yousefi, U. Kostiv, D. Horak, N. Pourreza, A. Merkoçi, ACS Nano **2015**, 9:7296

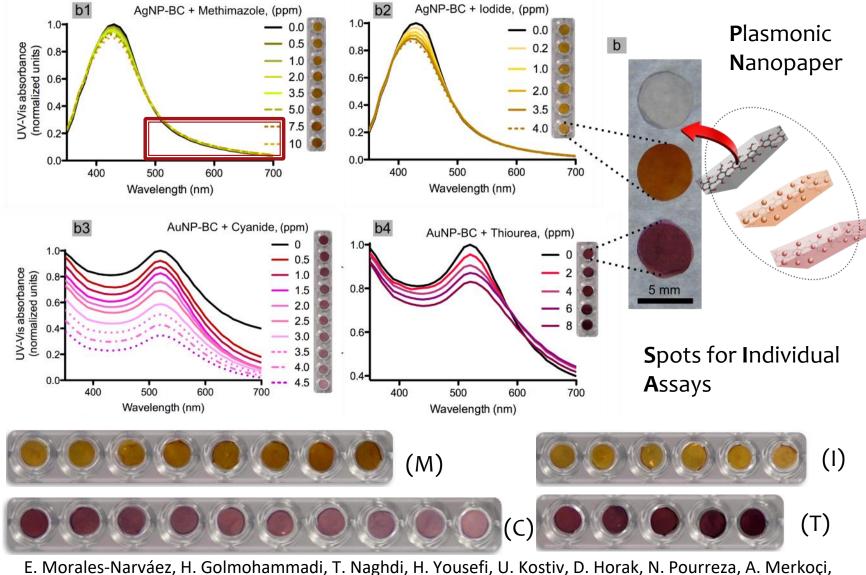
Nanopaper as an Optical Sensing Platform



E. Morales-Narváez, H. Golmohammadi, T. Naghdi, H. Yousefi, U. Kostiv, D. Horak, N. Pourreza, A. Merkoçi,

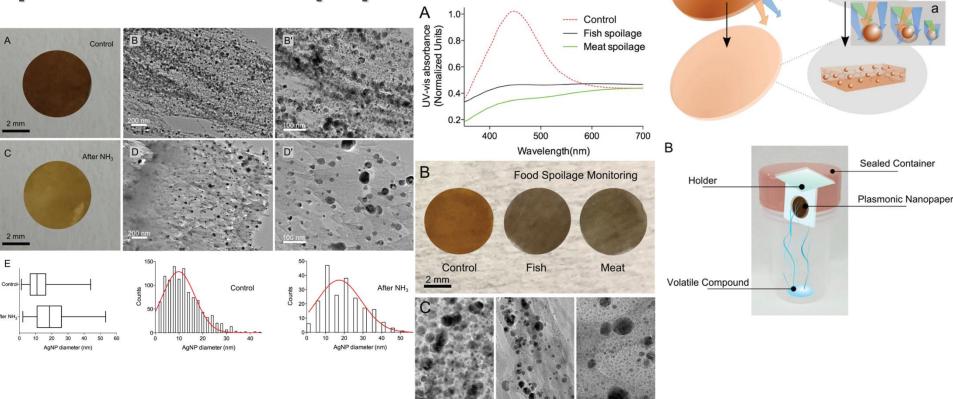
ACS Nano 2015, 9:7296

Nanopaper as an Optical Sensing Platform



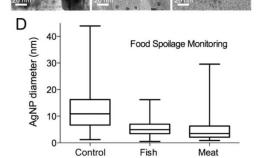
<u>orales-Narváez</u>, <u>H. Golmohammadi</u>, T. Naghdi, H. Yousefi, U. Kostiv, D. Horak, N. Pourreza, A. Merkoçi, ACS Nano **2015**, 9:7296

Visual detection of volatile compounds in a piece of AgNP-based plasmonic nanopaper



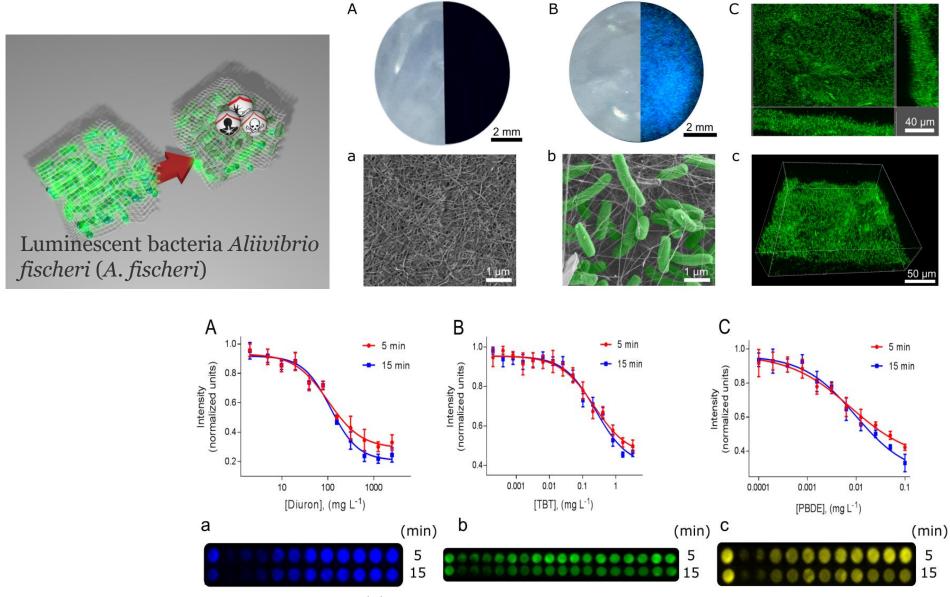
Modulation of population density and size of **silver nanoparticles** embedded in bacterial cellulose via ammonia exposure:

Merkoçi et al., Nanoscale, 2016, 8, 7984



a simple visual detection, which opens the way to innovative approaches and capabilities in gas sensing and smart packaging

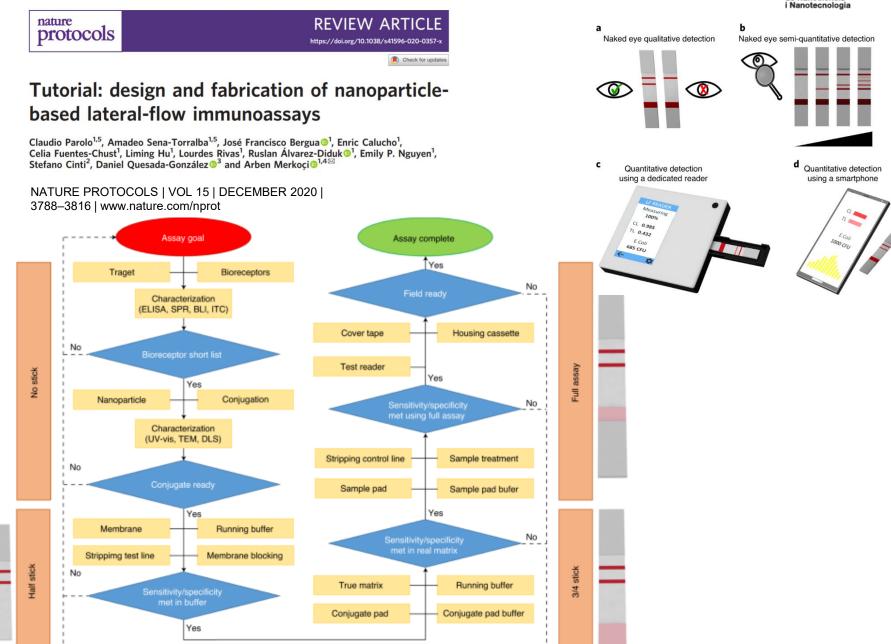
Bioluminescent Nanopaper for the rapid Screening of Toxic Substances



Merkoçi et al., NanoResearch, 2018, Vol 11 (1), pp 114-125

NP-based lateral flow immunoassay





paperdrOpdx

A small-medium enterprise founded in 2016



Spin-off company of:





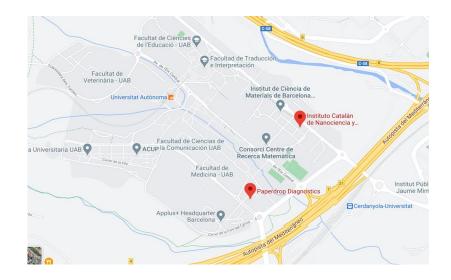
Barcelona Institute of Science and Technology

BIST

Part of:







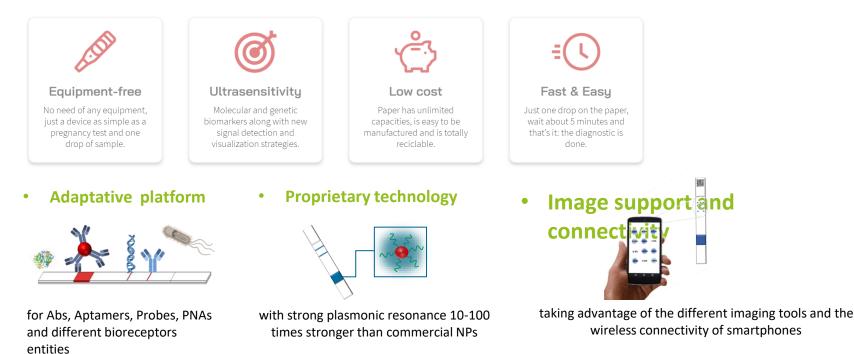


Professional & Self-testing In Vitro Diagnostics



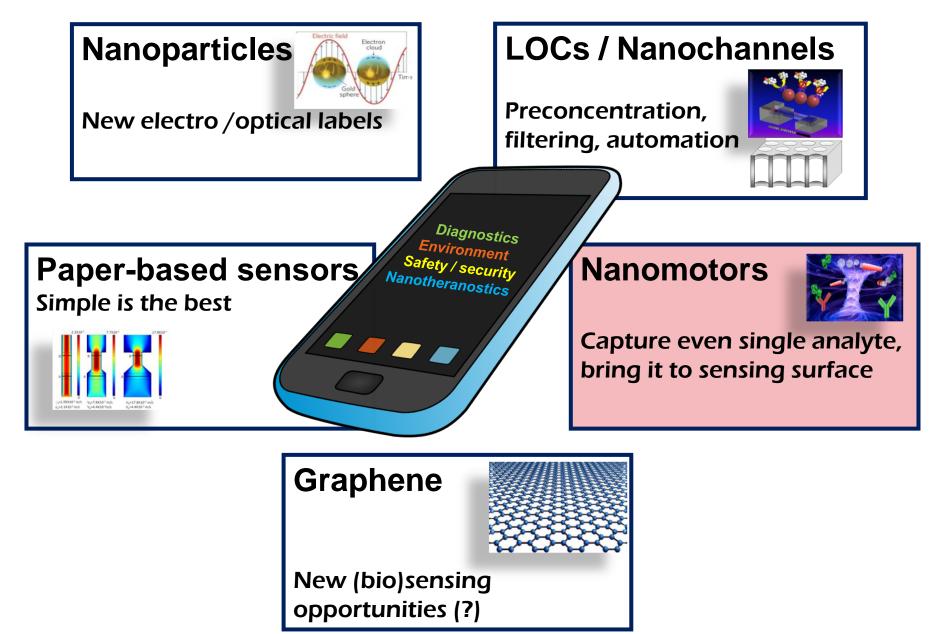
Technology awarded with the Seal of Excellence of the European Commission

Rapid diagnostic tests based on paper-microfluidics highly flexible that enable the integration of a wide range of bioreceptors in a very short period of time.



OUR MOTIVATION

To design new simple nanobiosensors and improve existing ones



Nano/micromotors (Collaboration with J.Wang, USA Maria Guix

Target Bacteria

Microsubmarines for Effective Removal

ACS Nano, 2012, 6, 4445-4451

Nanoscale, 2013

of Oil

SAM-modified microsubmarine Lectin

Oil droplets

Bacterial Isolation by Lectin-Modified Enhancing of biosensing Microengines **Template-based catalytic microengines** (no need for clean room) Au/Ni/PANI Polyaniline/Pt 2 µm В Nano Letters, 12, 396-401. 2012 Α Superhydrophobic Alkanethiol-Coated

 $2H_2O_2(I) \rightarrow 2H_2O(I) + O_2$

6

500 nm

Magnetic Control Trilaver PANI/Ni/Pt Microengine

Coupling nanomotors effect with biosensing

Bacterial Isolation by lectin-modified microengines

Selective capture of *Escherichia coli* from **food** and **clinical** samples

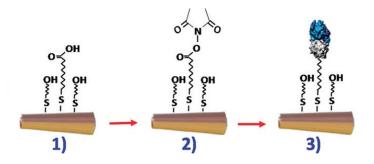
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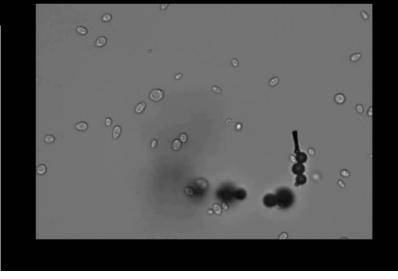
Drinking water



Selective pick-up of E. coli in presence of S. cerevisiae

Template-based self-propelled microengine Au/Ni/PANI/Pt Modified with Concanavalin A (ConA: lectin bioreceptor)

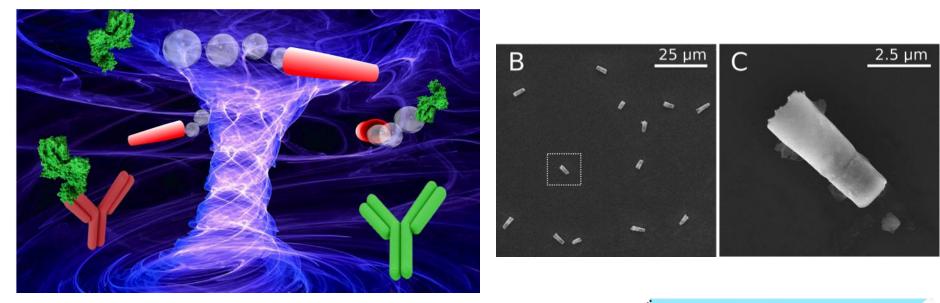


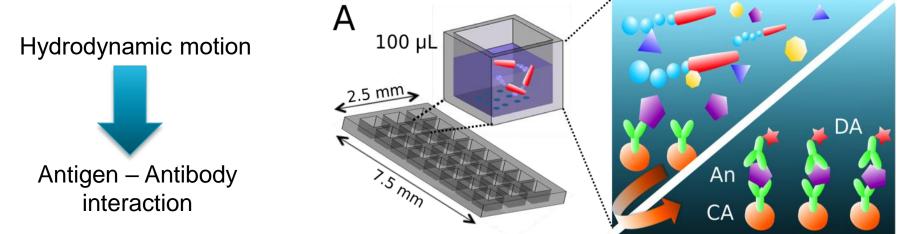


S. Campuzano et al. Nano Lett. 12, 2012, 396-401.



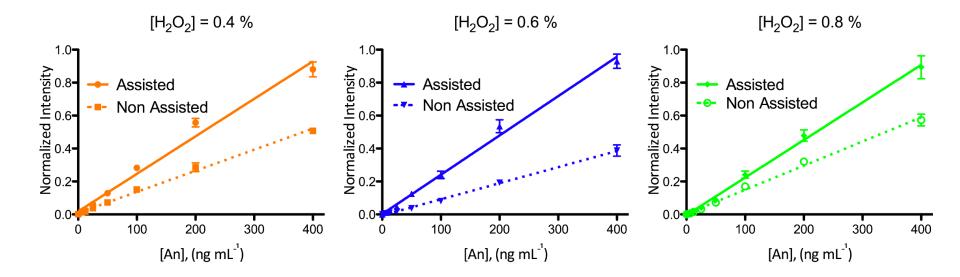
Microengines meet µA technology

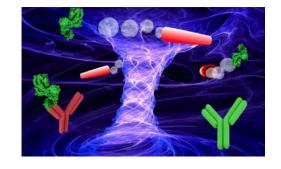




Morales-Narváez, Guix*, Medina-Sánchez, Mayorga-Martinez, Merkoçi, Small, 2014

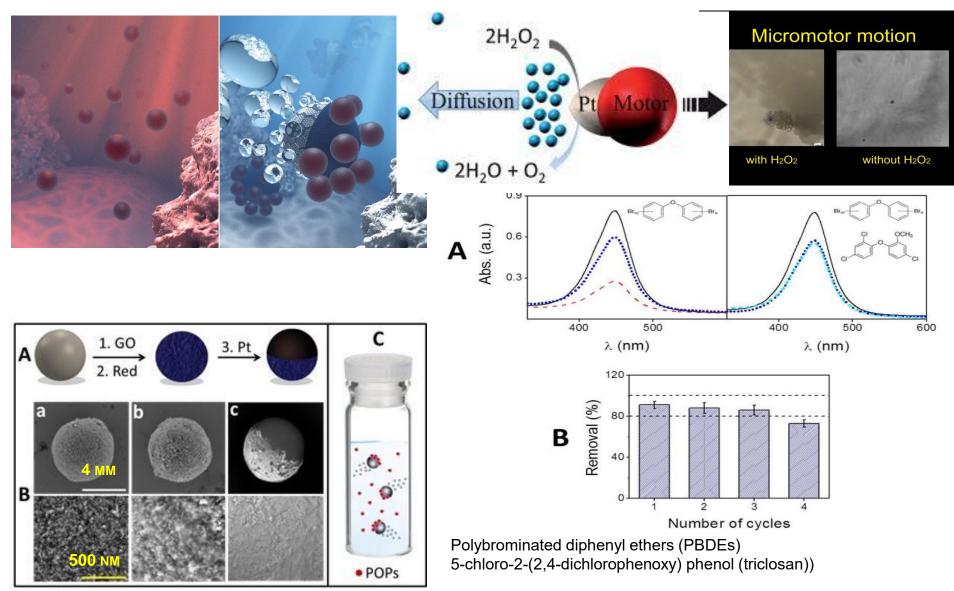
Microengines meet µA technology





Morales-Narváez*, Guix*, Medina-Sánchez, Mayorga-Martinez, Merkoçi, Small, 2014

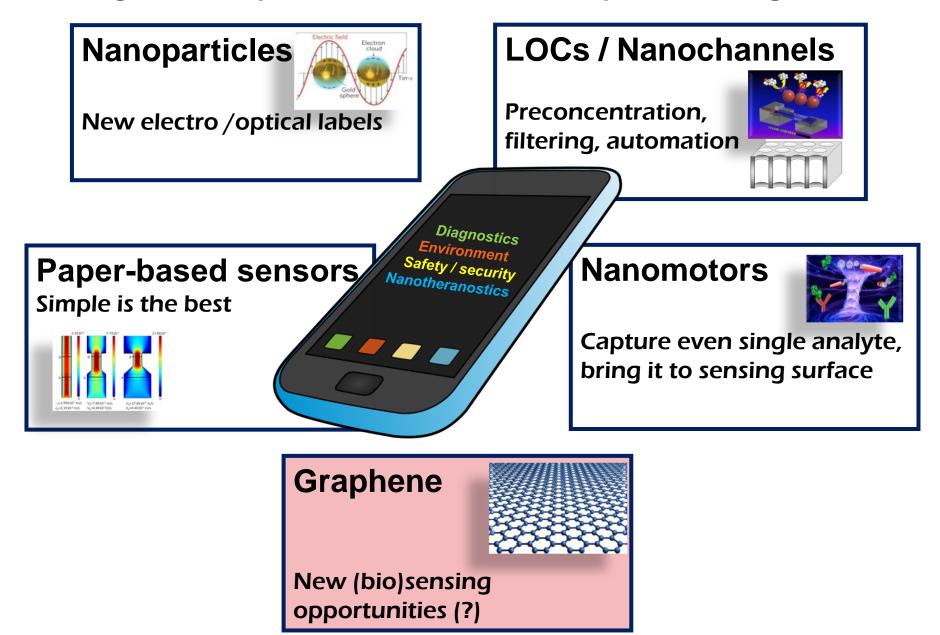
SiO₂@rGO-Pt Janus Micromotors Enhanced removal of Persistent Organic Pollutants (POPs)



Jahir Orozco, Luiza A. Mercante, Roberto Pol, Arben Merkoçi, J. Mater. Chem. A, 2016,4, 3371-3378

OUR MOTIVATION

To design new simple nanobiosensors and improve existing ones

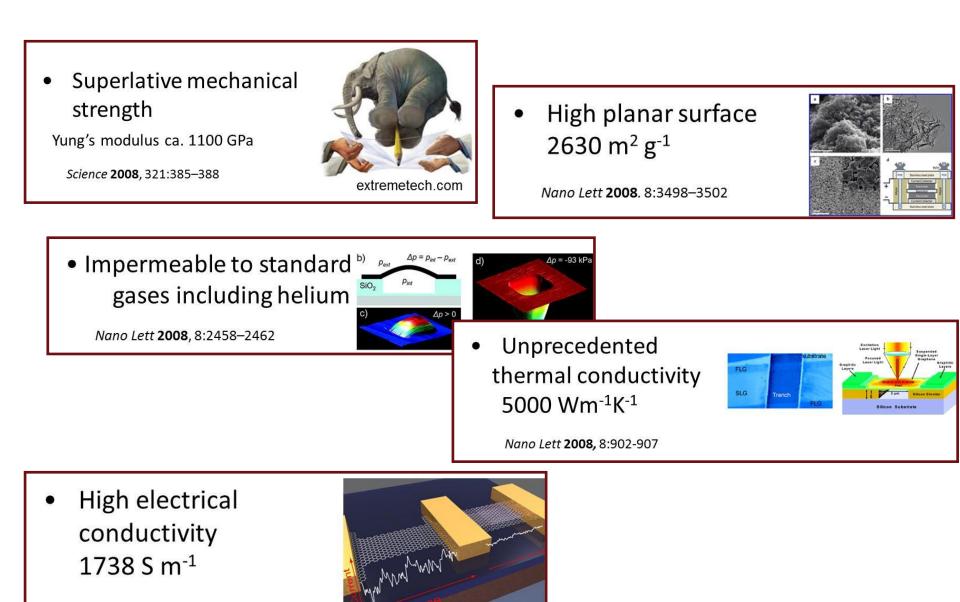








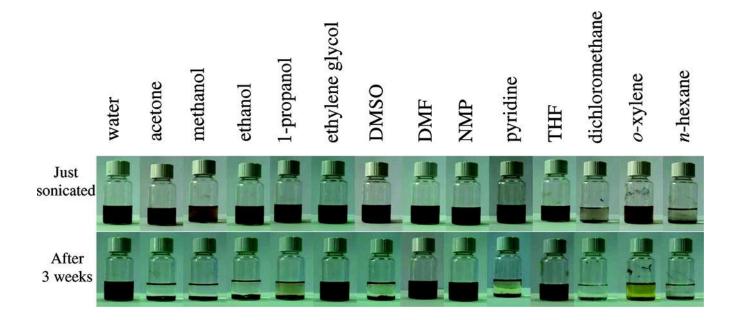




Science 2012, 335:1326-1330



GO can be processed in suspension form

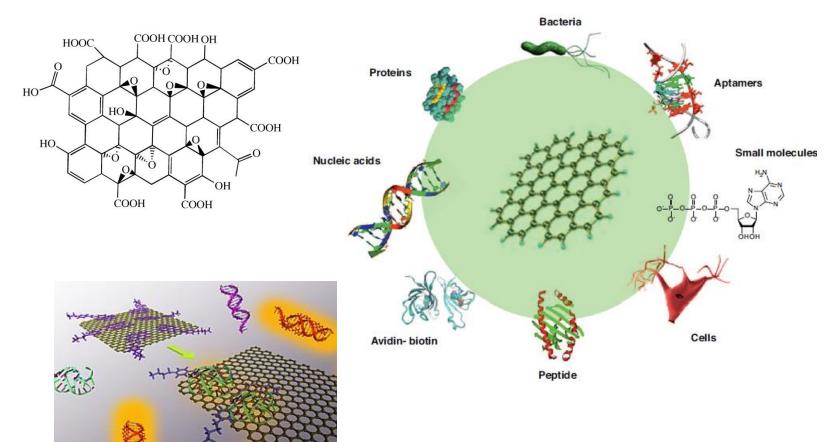




Langmuir **2008**, 24:10560-10564



• Direct wiring with biomolecules



Chem Soc Rev 2010, 39:4146-4157

Trends Biotechnol **2011**, 29:205-212





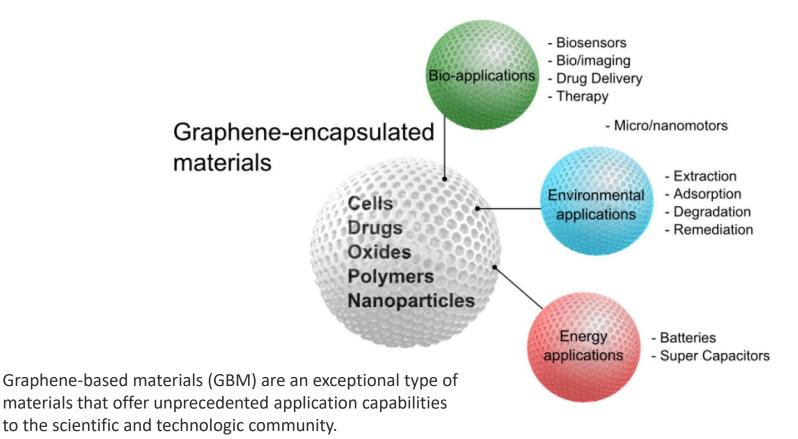
Progress in Materials Science

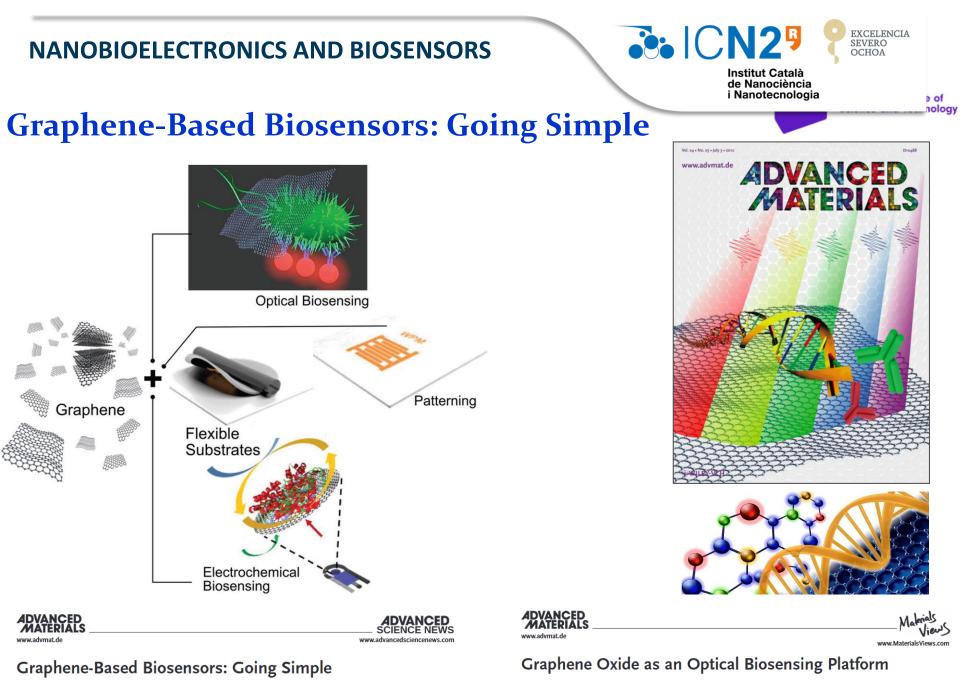
Volume 86, May 2017, Pages 1-24



Graphene-encapsulated materials: Synthesis, applications and trends

Eden Morales-Narváez ª, Lívia Florio Sgobbi ª^{, b}, Sergio Antonio Spinola Machado ^b, Arben Merkoçi ^{a, c} Ӓ 🖾



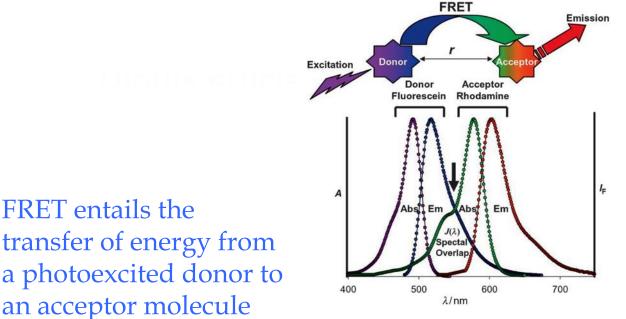


Eden Morales-Narváez, Luis Baptista-Pires, Alejandro Zamora-Gálvez, and Arben Merkoçi*

Eden Morales-Narváez and Arben Merkoçi*



Excellent quencher of fluorescence



Angew Chem Int Ed 2006, 45:4562 – 4588

Typically $E = 1 / [1 + (R / R_0)^6]$ d ~ 10 nm **Using Graphene** $E = 1 / [1 + (R / R_0)^4]$ d ~ 30 nm

J Chem Phys **2009**, 130:086101

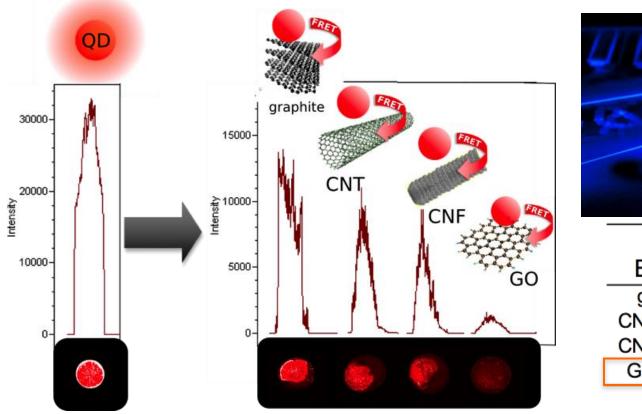


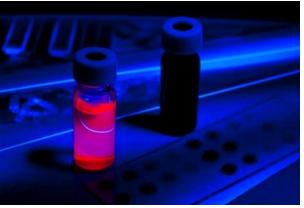
FRET entails the

Graphene Oxide in FRET (solid phase)



Simple FRET evidence for the ultrahigh QD quenching efficiency by GO compared to other carbon structures

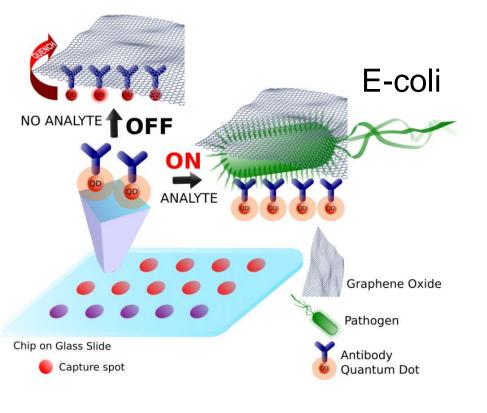


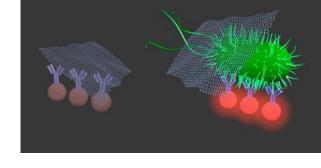


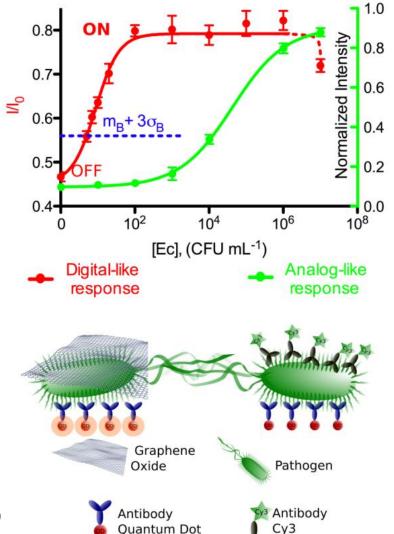
QD Quenching	
Efficiency range (%)	
g	17±05 ÷ 66±17
CNT	63±07 ÷ 71±01
CNF	52±10 ÷ 74±07
GO	91±02 ÷ 97±01

Morales-Narváez, Merkoçi, et al. Carbon 2012, 50:2987

GO as a pathogen-revealing agent







Patent

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

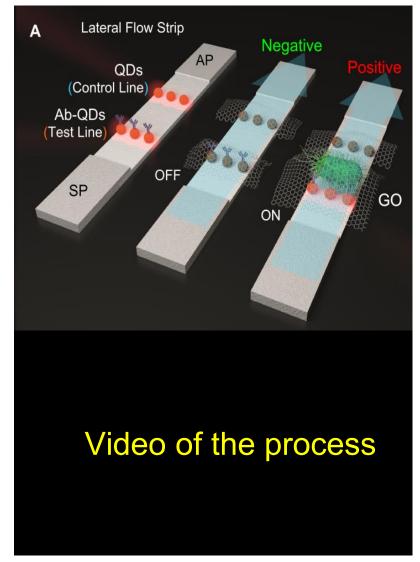
(19) World Intellectual Property Organization International Bureau

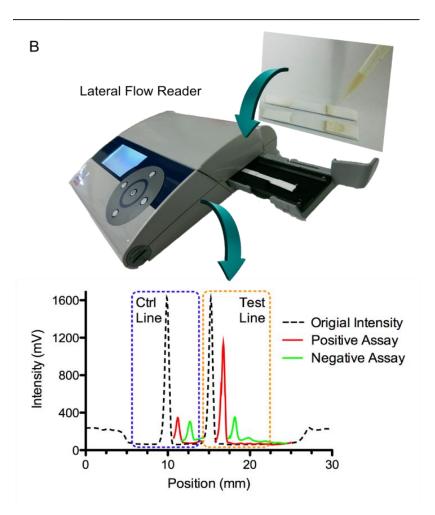
(43) International Publication Date 23 April 2015 (23.04.2015) WIPOIPCT

(10) International Publication Number WO 2015/055708 A1

Morales-Narváez, Merkoçi, et al., Angew. Chem. Int. Ed. 2013, 52, 13779

GO as a pathogen-revealing agent





Patent

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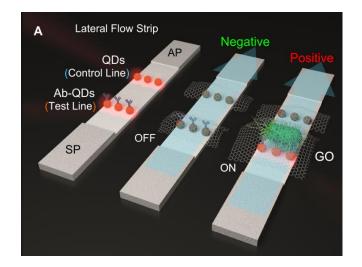
(43) International Publication Date 23 April 2015 (23.04.2015) WIPO | PCT

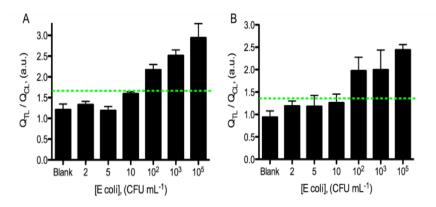
(10) International Publication Number WO 2015/055708 A1

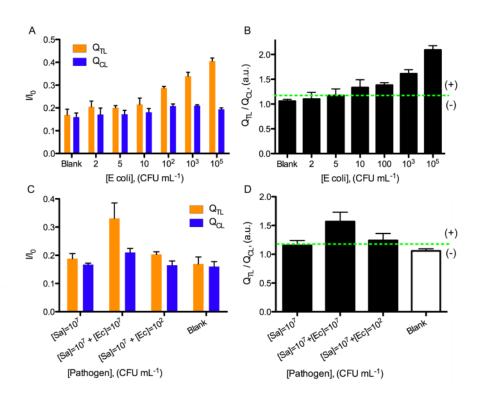
ales-Narváez, Naghdi, Zor, Merkoçi, Anal. Chem. 2015, 87:8573



GO as a pathogen-revealing agent

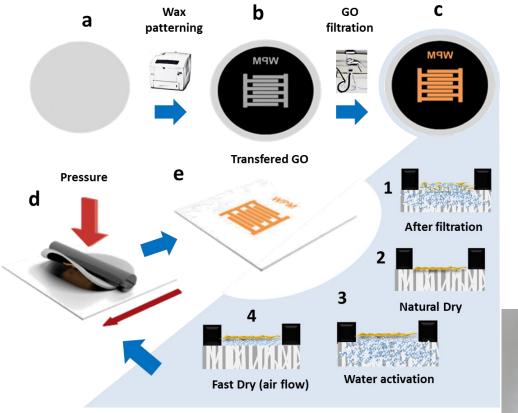








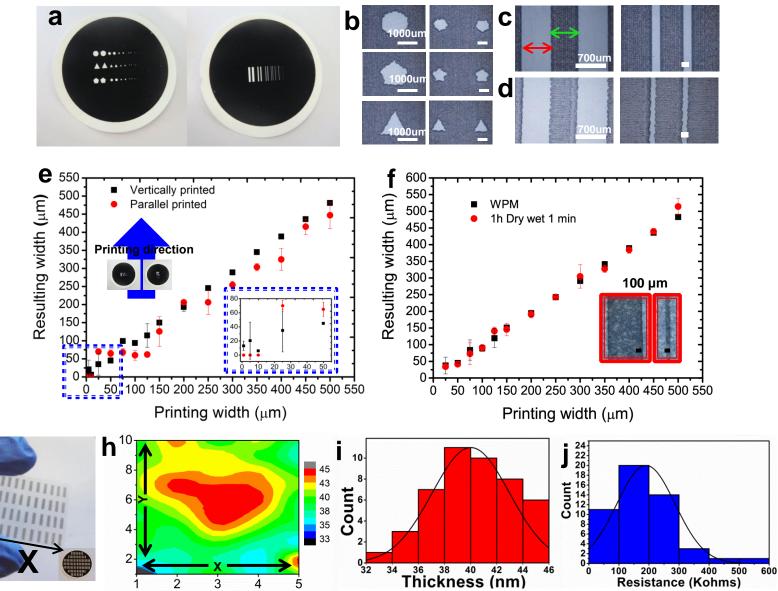
Water Activated Graphene Oxide Transfer Using Wax Printed Membranes for Fast Patterning of Electrical Devices





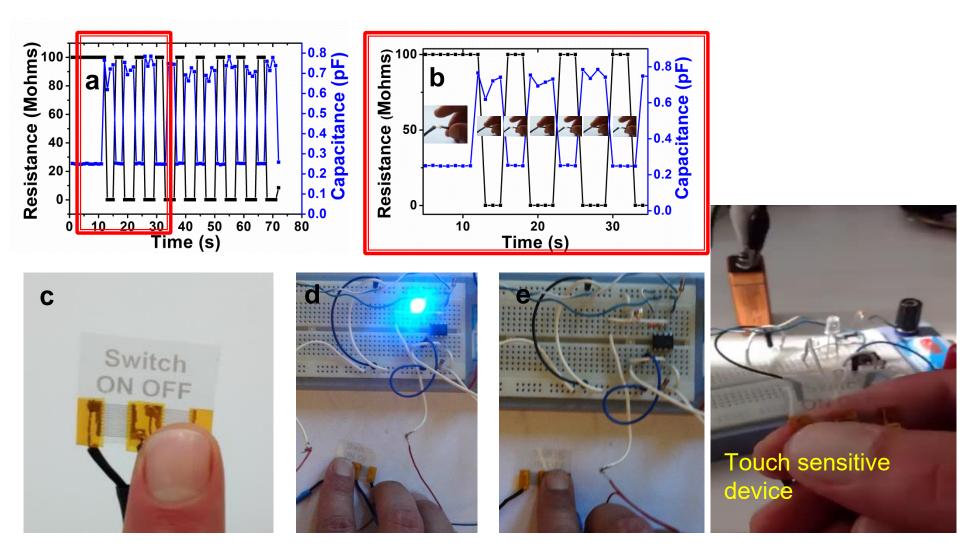
Baptista-Pires, Mayorga-Martínez, Medina-Sánchez, Montón, Merkoçi, ACS Nano 2016, 10, 853-860

Water Activated Graphene Oxide Transfer Using Wax Printed Membranes for Fast Patterning of Electrical Devices



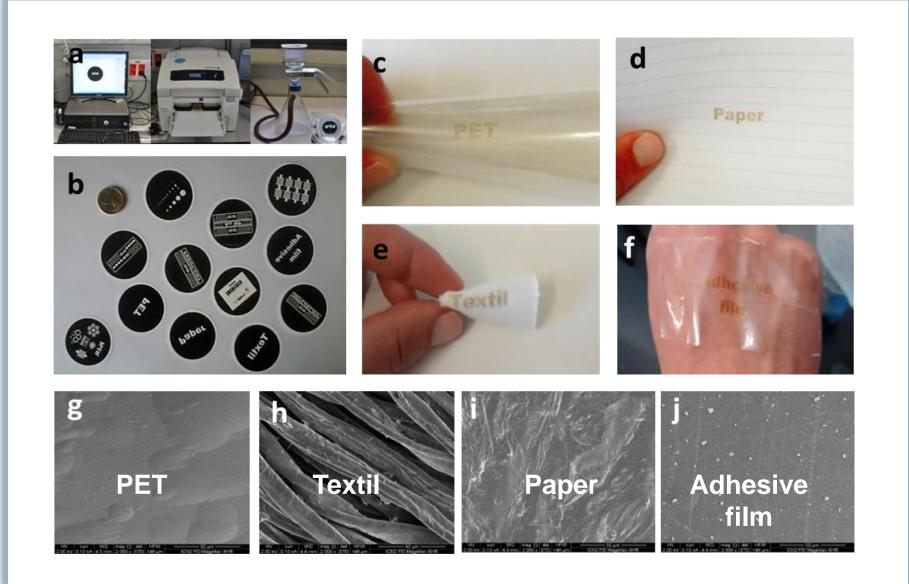
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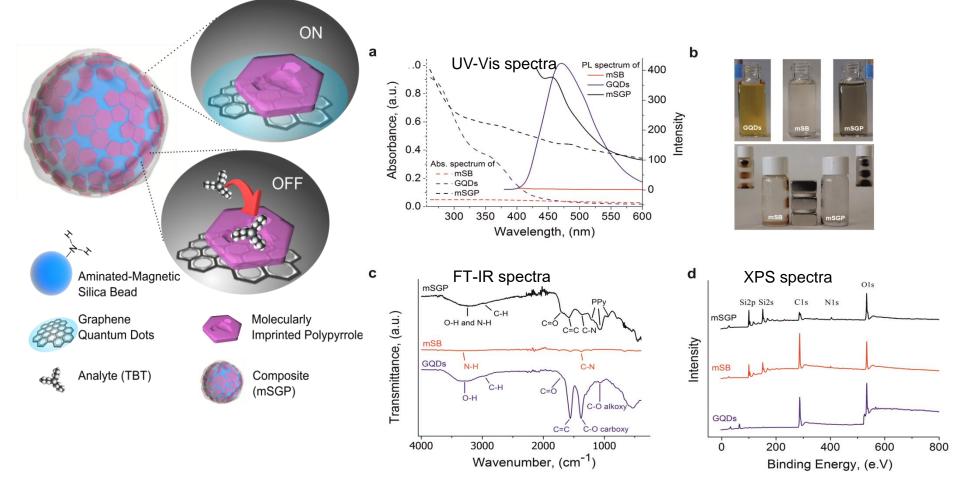
Baptista-Pires, Mayorga-Martínez, Medina-Sánchez, Montón, Merkoçl, ACS Nano 2016, 10, 853-860

Transfer onto flexible substrates



GO as a PL donor

Graphene Quantum Dots-based Photoluminescent Sensor: A Multifunctional Composite for Pesticide Detection

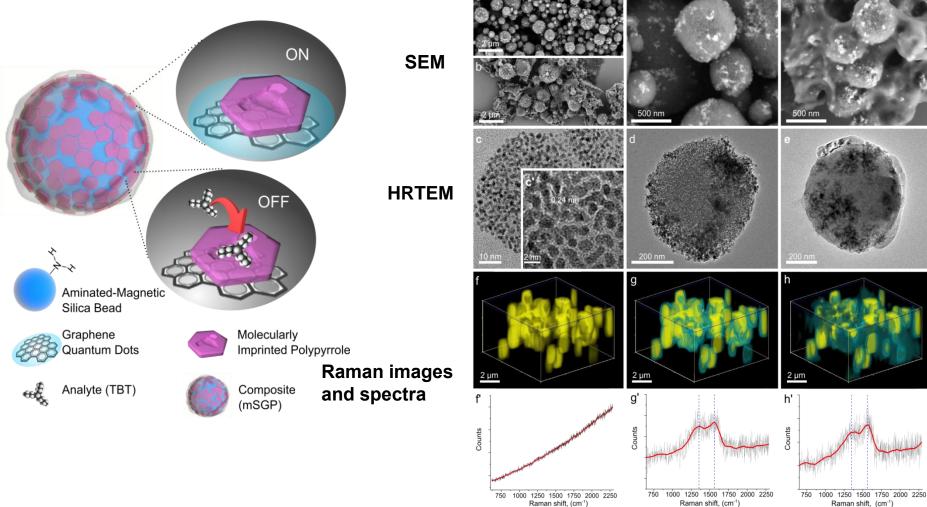




Zor, Morales-Narváez, Zamora-Gálvez, Bingol, Ersoz, Merkoçi, ACS Appl Mater Interfaces. 2015, 7:20272

GO as a PL donor

Graphene Quantum Dots-based Photoluminescent Sensor: A Multifunctional Composite for Pesticide Detection

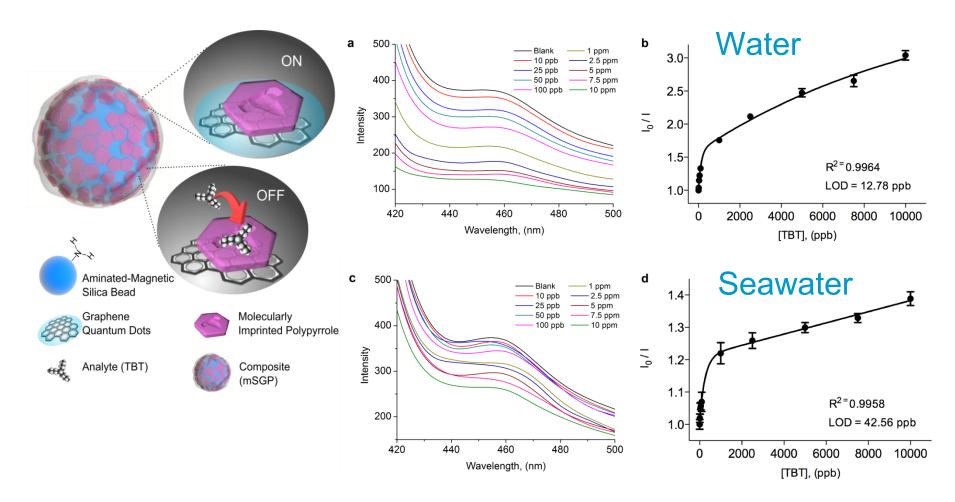




Zor, Morales-Narváez, Zamora-Gálvez, Bingol, Ersoz, Merkoçi, ACS Appl Mater Interfaces. 2015, 7:20272

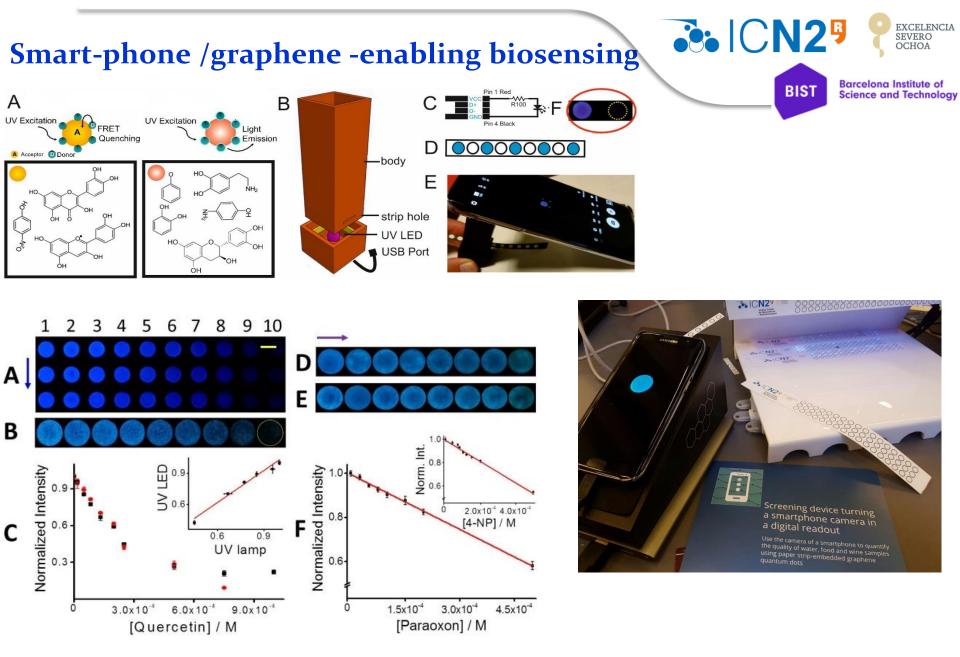
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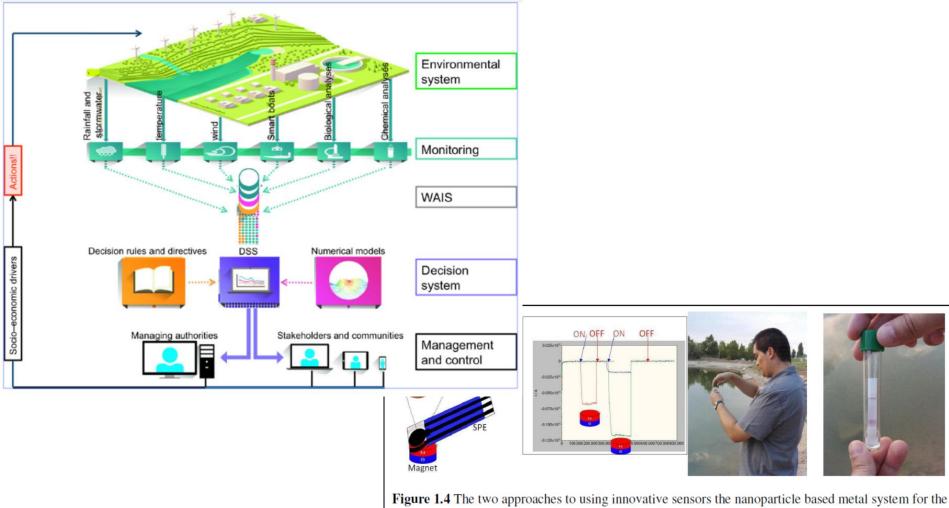
Smart-phone paper/graphene-based sensor for safety and security applications European Patent Application P30540EP00

Arben Merkoçi et al. Scientific Reports, 2017,





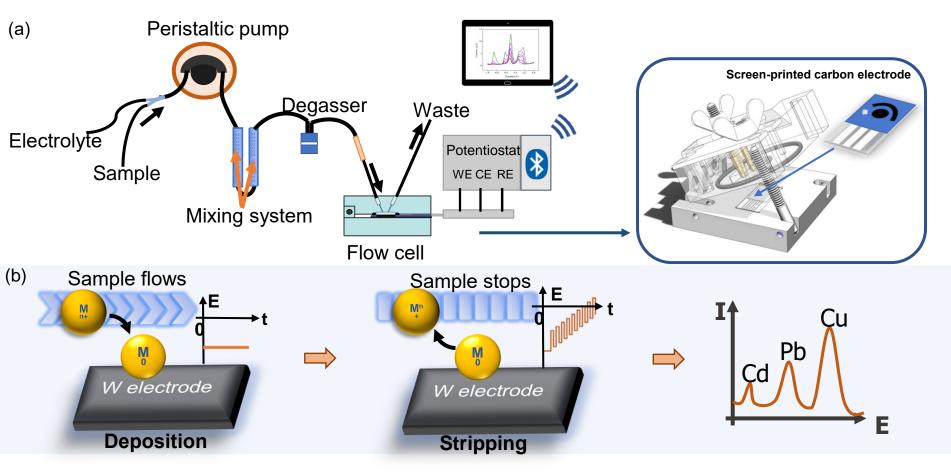
Innovative monitoring tools for river and lake water quality, and a new business model for 2020 and beyond



boats (left), and the land-based strips (right)



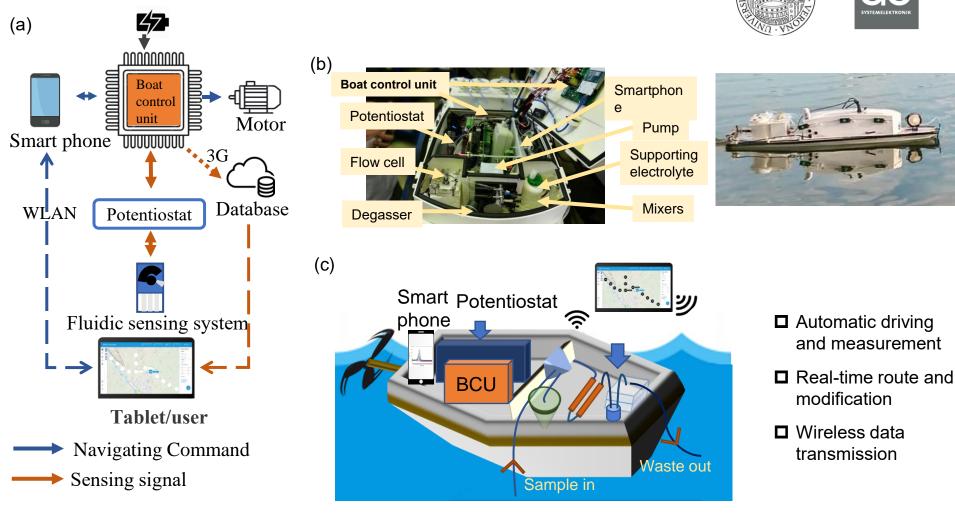
Laboratory set-up for *in-situ* and automatic heavy metal sensing measurements



Yang, Qiuyue, Merkoçi et al. "Development of a heavy metal sensing boat for automatic analysis in natural waters utilizing anodic stripping voltammetry." ACS ES&T Water 1.12 (2021): 2470-2476.



Autonomous boat for heavy metal sensing measurements



Yang, Qiuyue, Merkoçi et al. "Development of a heavy metal sensing boat for automatic analysis in natural waters utilizing anodic stripping voltammetry." ACS ES&T Water 1.12 (2021): 2470-2476.

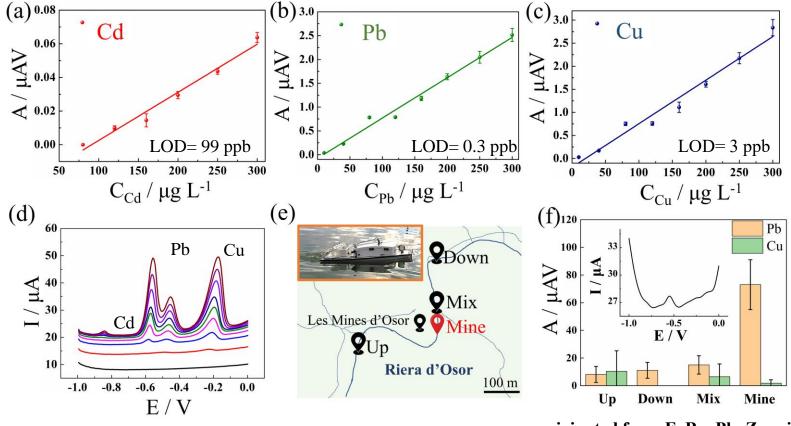


Heavy metal sensing performance





University of Natural Resources and Life Sciences, Vienna



originated from F–Ba–Pb–Zn mine vein

Yang, Qiuyue, Merkoçi et al. "Development of a heavy metal sensing boat for automatic analysis in natural waters utilizing anodic stripping voltammetry." ACS ES&T Water 1.12 (2021): 2470-2476.



Navigating performance







University of Natural Resources and Life Sciences, Vienna



Conclusions



Nanobiosensors represent a great alternative for environment monitoring

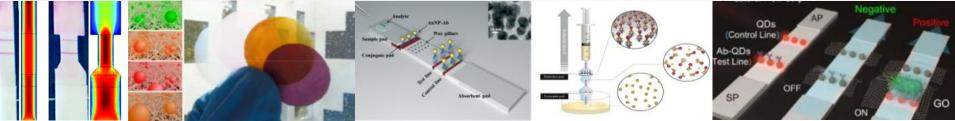
These devices and related fabrication technologies offer unpresented alternative for the democratization of monitoring systems

Nanomatarials can be easily coupled to paper-based platforms to build cost/efficient nanobiosensors

Nanomaterials exhibit unprecedented properties as either electrical or optical transducer for biosensing applications

Their properties and related platforms can enable:

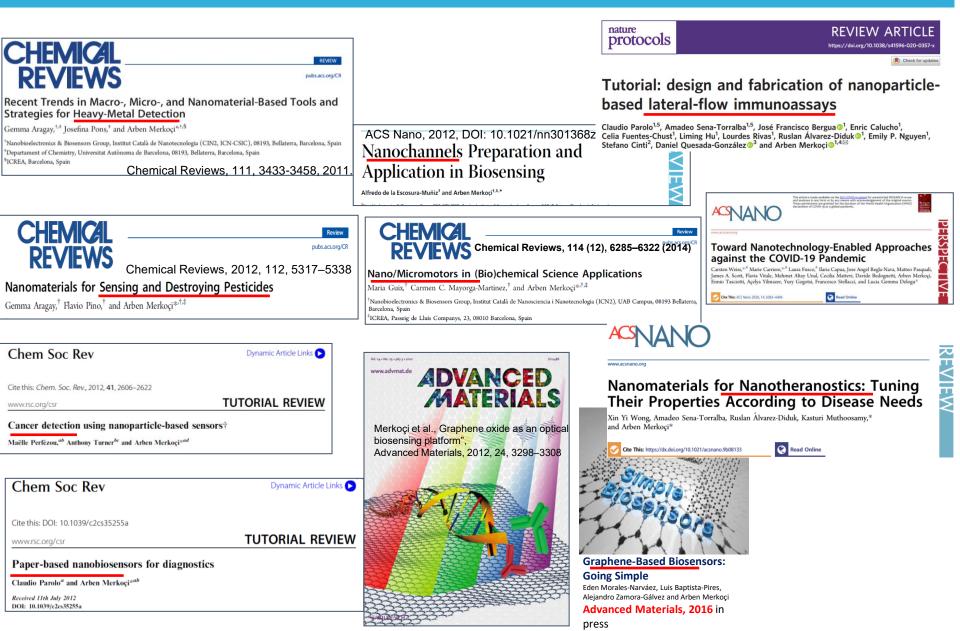
- Connection to a variety of (bio)receptors and nanomaterials
- Simple assay procedures and avoid time consuming labours
- Compatibility with mobile phone technology and other smart environment monitoring systems.



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Nanodiagnostics to Face SARS-CoV-2 and Future Pandemics: From an Idea to the Market and Beyond

Giulio Rosati, Andrea Idili, Claudio Parolo, Celia Fuentes-Chust, Enric Calucho, Liming Hu, Cecilia de Carvalho Castro e Silva, Lourdes Rivas, Emily P. Nguyen, José F. Bergua, Ruslan Alvárez-Diduk, José Muñoz, Christophe Junot, Oriol Penon, Dominique Monferrer, Emmanuel Delamarche, and Arben Merkoçi[®]



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Chemical Society Reviews

Attomolar analyte sensing techniques (AttoSens): a review on a decade of progress on chemical and biosensing nanoplatforms

Sruthi Prasood Usha, 🔞 a Hariharan Manoharan, a Rehan Deshmukh, a

Ruslan Álvarez-Diduk, 10 ^b Enric Calucho, ^b V. V. R. Sai 10 * ^a and Arben

Merkoçi 🗊 * bc



Trends in Chemistry

CellPress

Available online 25 April 2022

In Press, Corrected Proof (?)

Paper-based biosensors for cancer diagnostics

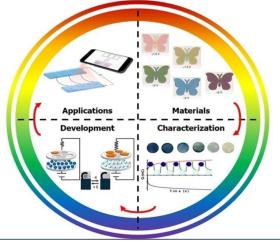
Claudia Pereira ^{1, 2, 3, 9}, Claudio Parolo ^{1, 4, 9}, Andrea Idili ^{1, 5}, Roger R. Gomis ^{6, 7}, Lígia Rodrigues ^{3, 8}, Goreti Sales ², Arben Merkoçi ^{1, 7} A 🛛

materialstoday

Electrochromism: An emerging and promising approach in (bio)sensing technology

Mohammad Amin Farahmand Nejad ^{1,2,4}, Saba Ranjbar ^{1,4}, Claudio Parolo ¹, Emily P. Nguyen ¹, Ruslan àlvarez-Diduk ¹, Mohammad Reza Hormozi-Nezhad ^{2,3}, Arben Merkoçi ^{1,4,}

Electrochromic sensors





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