Laboratoire de l'Intégration du Matériau au Système

JOURNEE OASU 12/10/17

Microcapteurs à ondes acoustiques pour la détection environnementale

Groupe Ondes – Equipe MDA

Présentée par O. TAMARIN

Hamida HALLIL, Jean Luc LACHAUD, Simon HEMOUR, Corinne DEJOUS, Dominique REBIERE

Sommaire

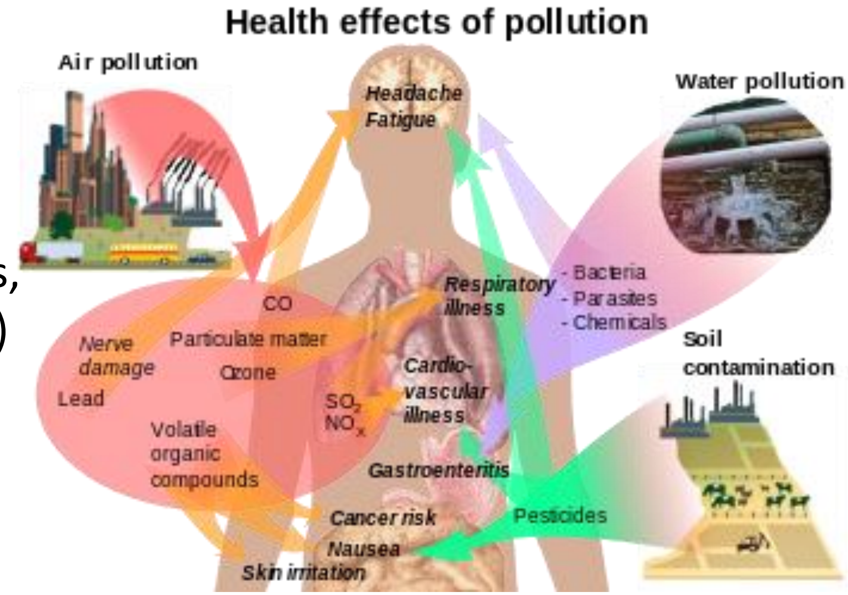
- **Introduction**
- **Plateforme de détection acoustique – quelques résultats**
- **Application à la détection de cibles de faible poids moléculaires**
- **Conclusion**

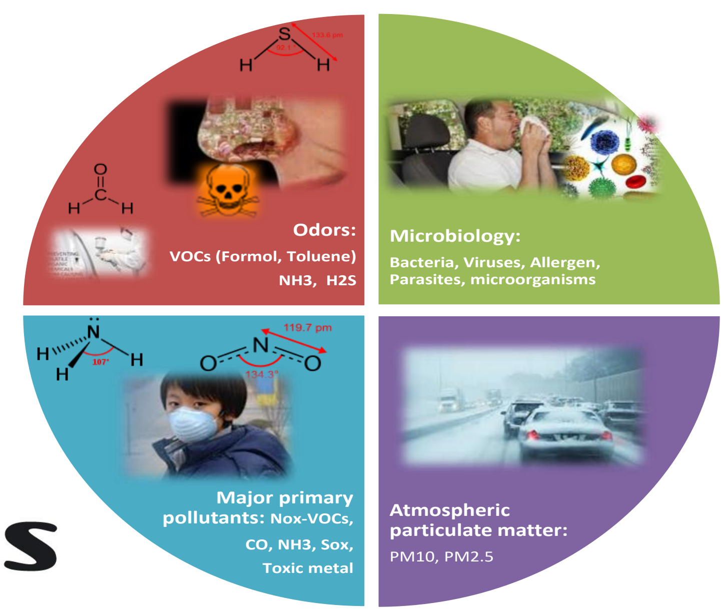
Current research themes

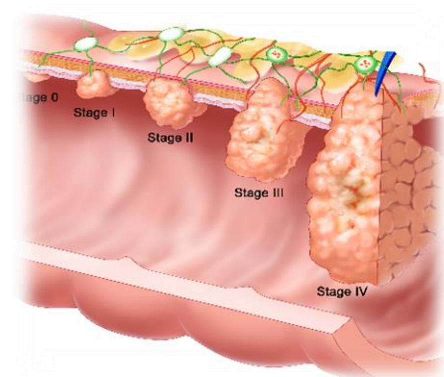
Applications in environmental and health areas:

- Detection in gaseous medium and in severe high temperature atmosphere : toxic compounds,
  VOC, ...& @ very low concentration, few ppb

- Detection in liquid medium (bacteria, virus, toxins, biomarkers, heavy metals and other target spices)
- Wireless implant communication
- **Using Microsensors to Promote Development of Innovative Therapeutic Nanostructures**

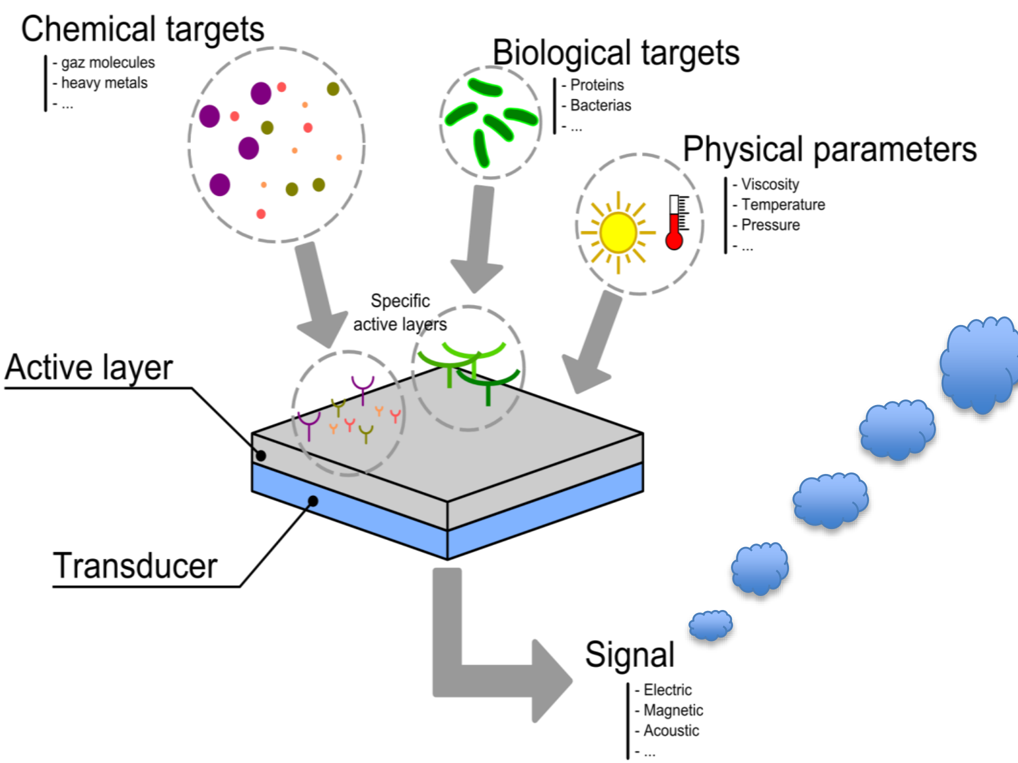




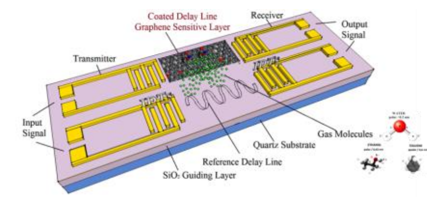


Ultrasensitive Acoustic & Electromagnetic & optical transducers : Application for gas and bio sensing

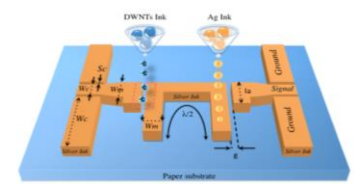
Gas or Biosensor principle



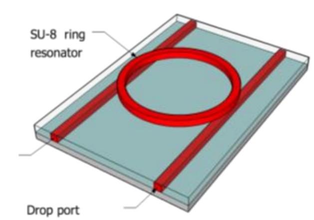
Acoustic platform
(piezoelectric substrate)



Electromagnetic platform
(flexible substrate)



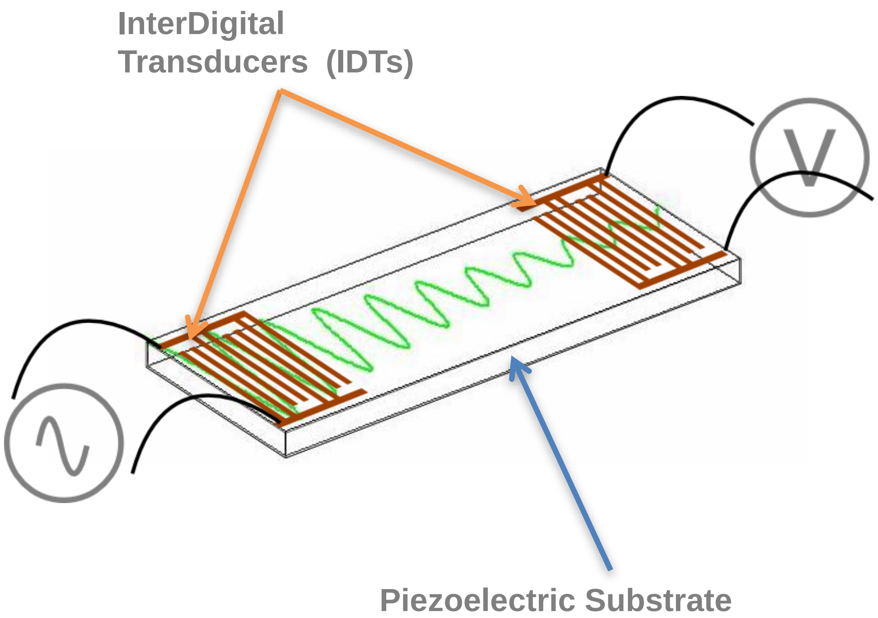
Optical platform
(polymer materials)



Ultrasensitive Acoustic transducer :
Application for gas and bio sensing

SAW Delay Line

- **Piezoelectric effect**
- Acoustic wave propagation using IDTs (Inter Digital Transducers)

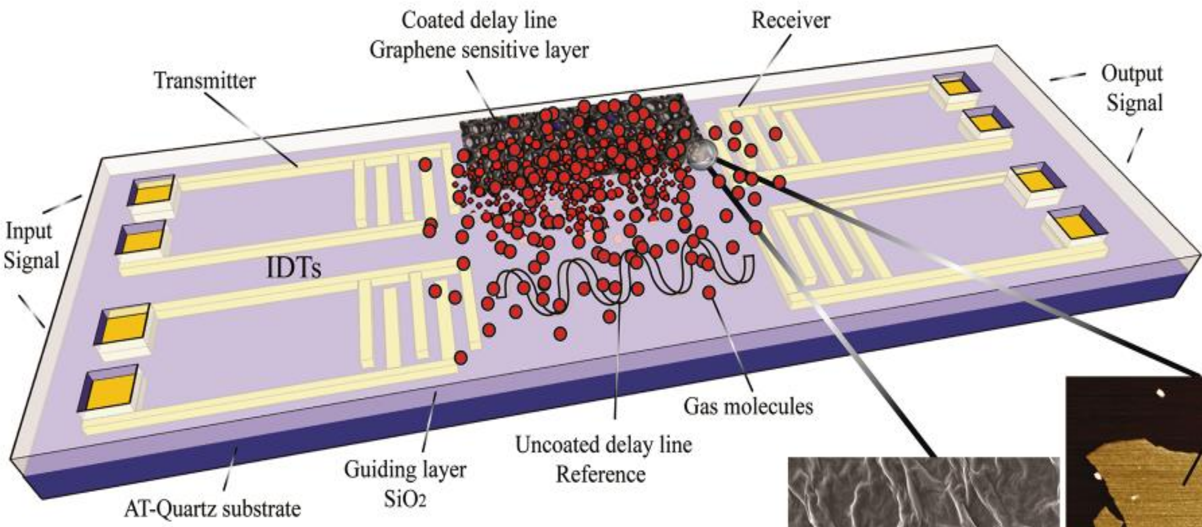


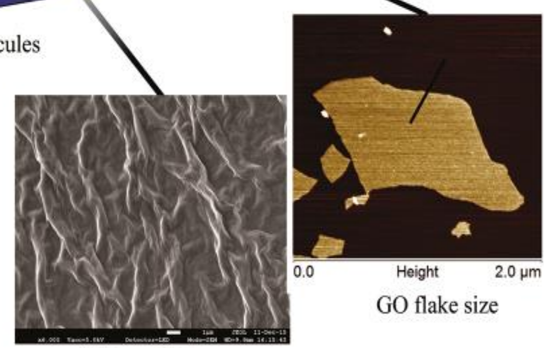
- Wave perturbation induces :
  - **Phase velocity variation**
  - **Attenuation (Insertion Loss)**

Ultrasensitive Acoustic transducer :
Application for gas and bio sensing

Love wave sensor

(A. E. H. **Love**, 1863-1940, Mathematician)





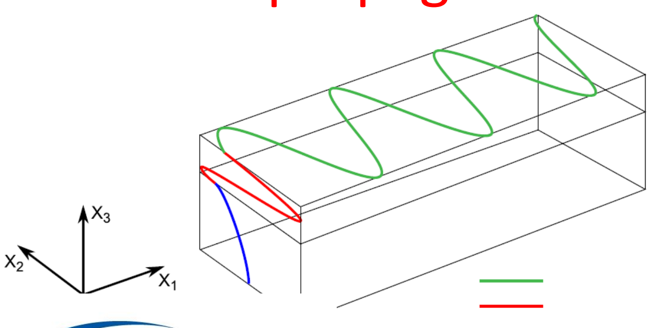
GO flake size

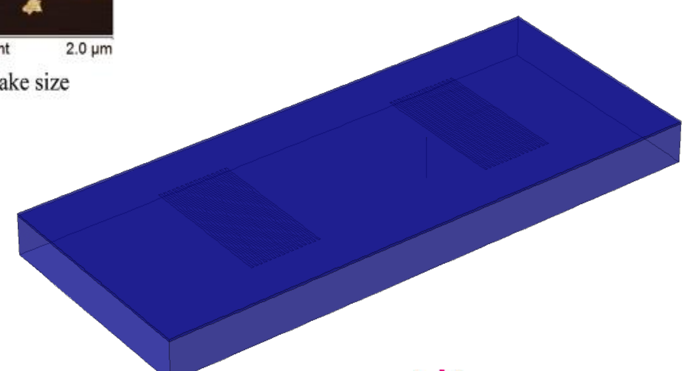
GO morphology

Love wave sensor advantages:

- High sensitivity in liquid media
- Real time detection
- Non invasive detection tool

Love wave propagation

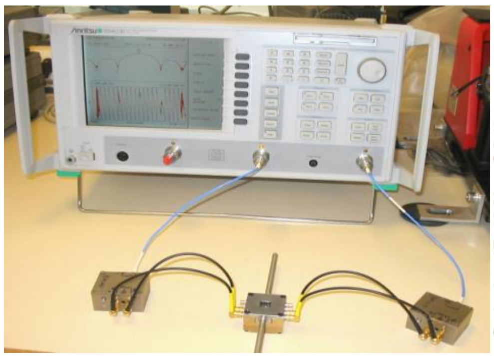




Ultrasensitive Acoustic transducer :

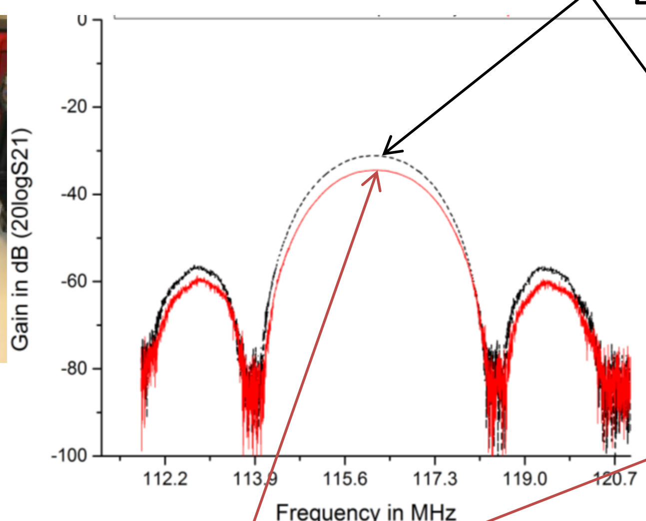
Application for gas and bio sensing

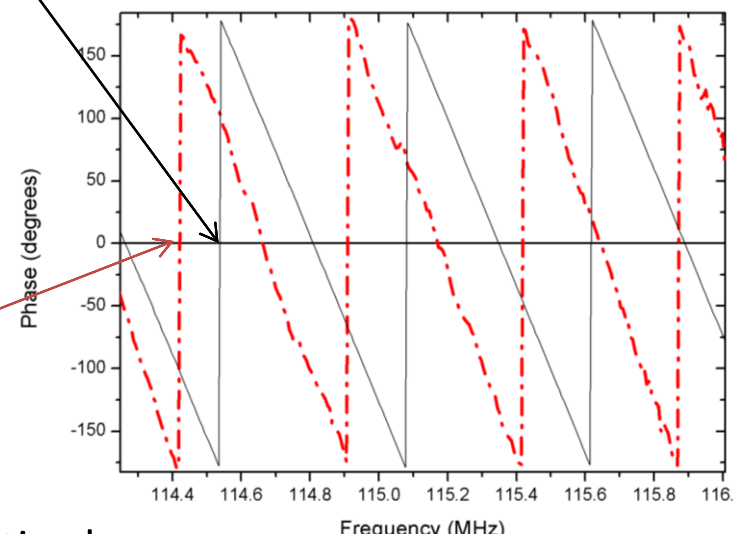
Sensor Electrical characterization



Vector Network Analyzer

- Before deposition of sensitive layer





- f_0 = 116.5 MHz
- Insertion Losses: -30 dB
- Delay: 1.542µs

$V_{phase\ velocity} \approx 4300 m.s^{-1}$

- After deposition of sensitive layer

insertion losses ~ 3.35 dB.

Phase: The frequency shift is ~ 97.5 KHz

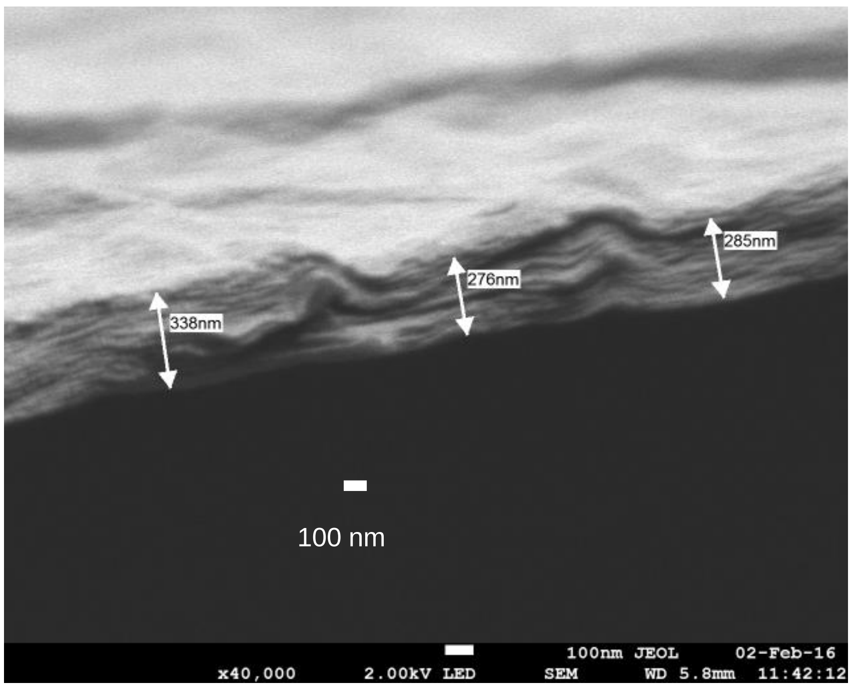
Ultrasensitive Acoustic transducer :

Application for gas sensing

Graphene oxide on Love wave sensor

Collaborative with advanced research Laboratories

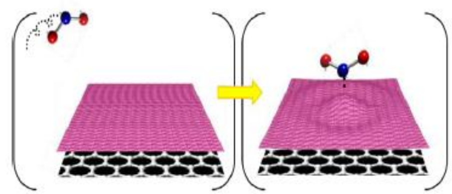
LAAS-CNRS Toulouse (France) | Instituto de Tecnología Química Valencia (Spain) | FORTH Institute of Electronic Structure and Laser Crete (Greece)



SEM images from the tilt-sectional areas of the GO sensing layer

Why Graphene as a sensitive layer?

F. Schedin et al., Nature 6, 652 (2007)



Highest Surface

2D material

Interactions on the surface

- **Gas molecules interactions**
  - Adsorption/desorption modifies the total mass of the sensitive layer
  - Insertion loss are induced → Sensitivity
  - Physical or Chemical sorption (tailoring properties of Graphene)

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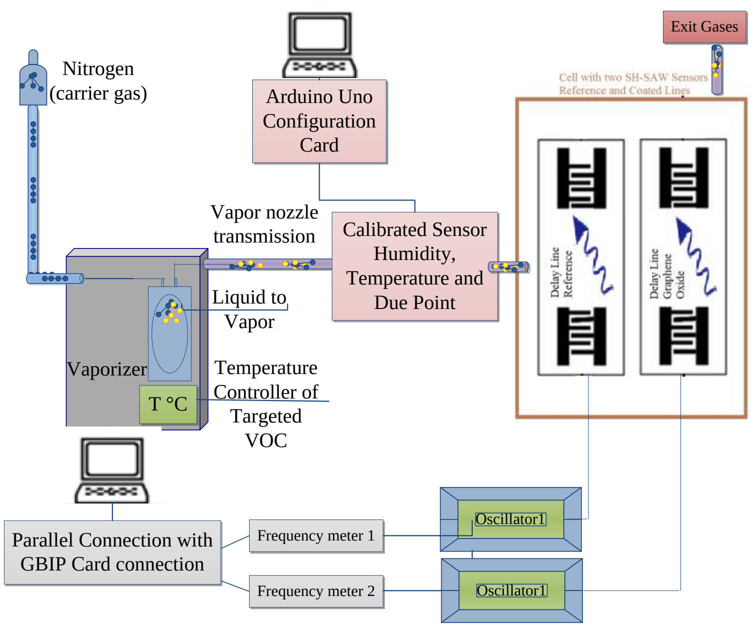
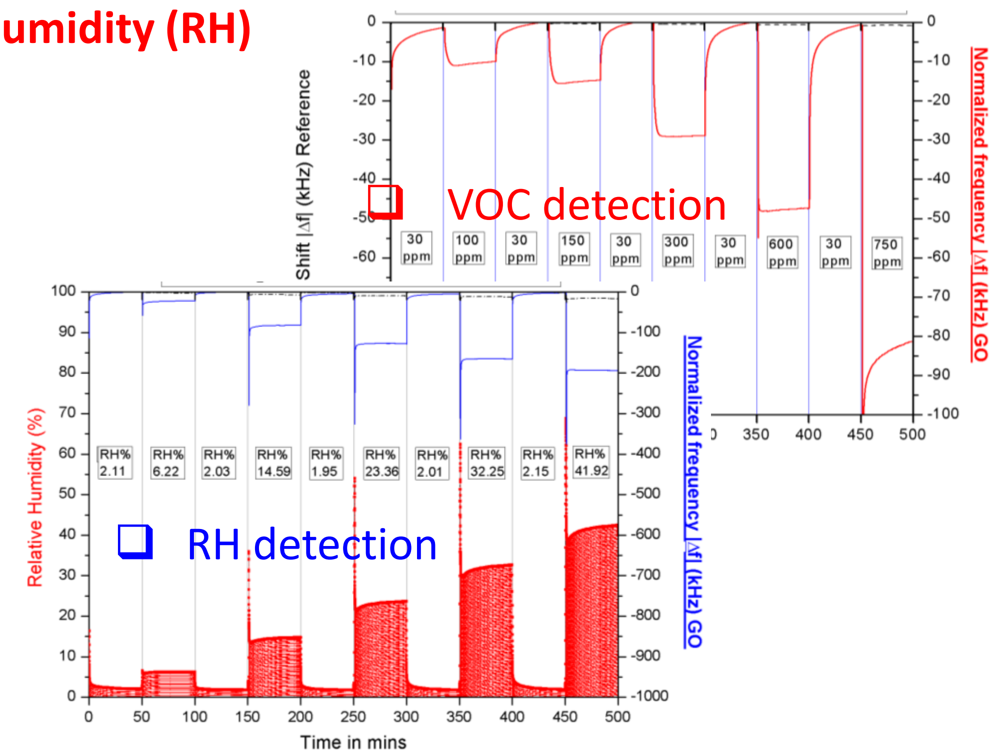
Ultrasensitive Acoustic transducer :
Application for gas sensing

Graphene oxide on Love wave sensor

Real time detection of Ethanol (VOC) and humidity (RH)





✓ Real time detection

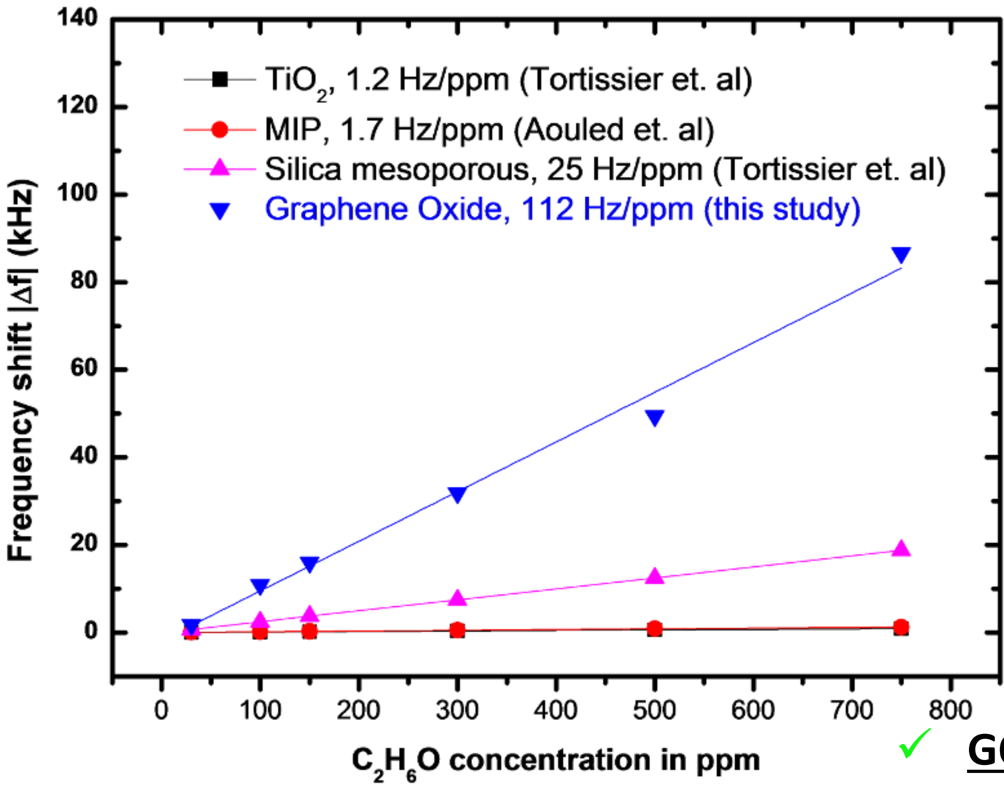
Nikolaou, I., Hallil, H., Plano, B., Deligeorgis, G., Conedera, V., Garcia, H., ... & Rebière, D. (2016). Drop-casted Graphene Oxide Love Wave sensor for detection of humidity and VOCs. *Journal of Integrated Circuits and Systems*, *11*(1), 49-56.

Ultrasensitive Acoustic transducer :

Application for gas sensing

Evaluation of our device based on graphene: Ethanol as VOC

Comparison of Graphene Oxide with relative functional materials under C_2H_6O vapors



Maximum sensitivity – 4 studies

- **GO efficiency**

- Silica mes. (4 times)
- MIP (50 times)
- TiO_2 (100 times)

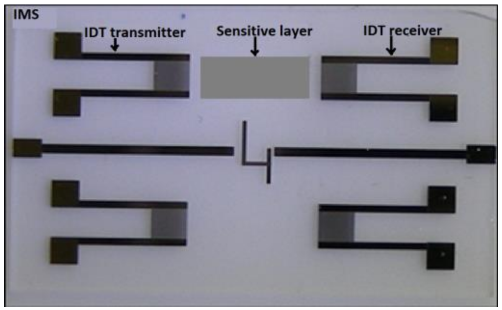
✓ **GO is promising** rather than alternative functional materials **(MIP, SiO_2, TiO_2)**

Nikolaou, I., Hallil, H., Conédéra, V., Deligeorgis, G., Dejous, C., & Rebiere, D. (2016). Inkjet–printed graphene oxide thin films on Love wave devices for humidity and vapor detection, J. IEEE Sensors.

Ultrasensitive Acoustic transducer :

Application for bio sensing

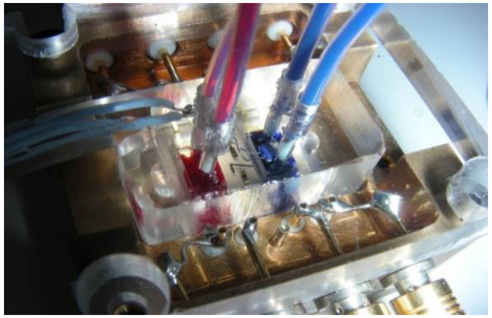
Test medium control for efficient detection:
Microfluidic chip renews the target species on the sensor surface

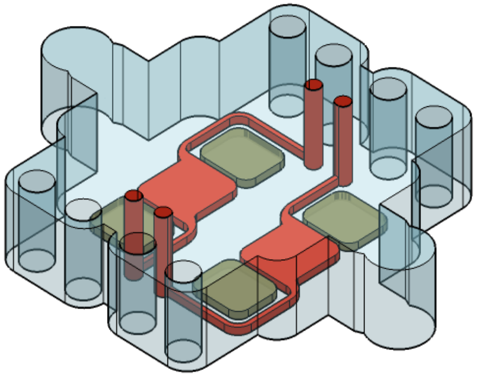


Microsystem (SH-SAW sensor)

Microfluidic chip

+

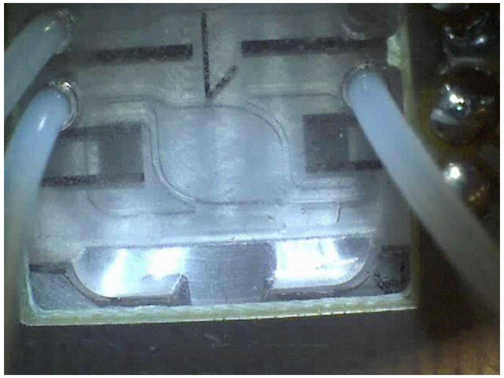


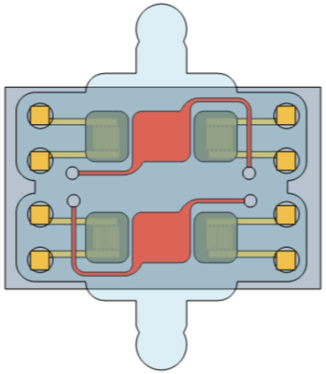


Microfluidic PDMS cell

- ✓ **Analysis chamber removable**
- ✓ **Biocompatible**

- ✓ **Real experimental conditions**
- ✓ **Low cost and easy to fabricate**

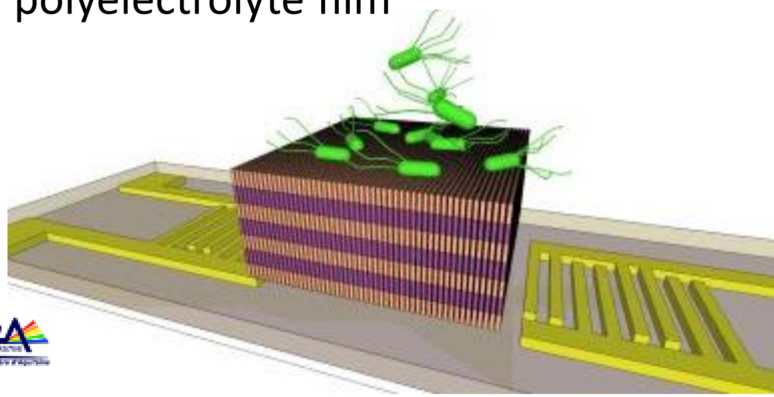


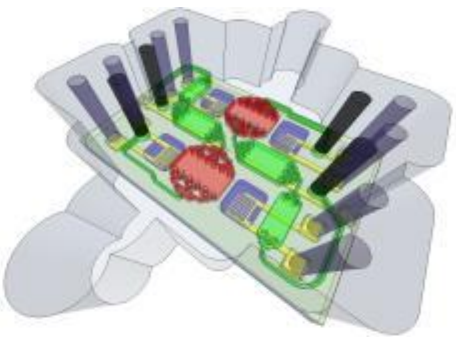


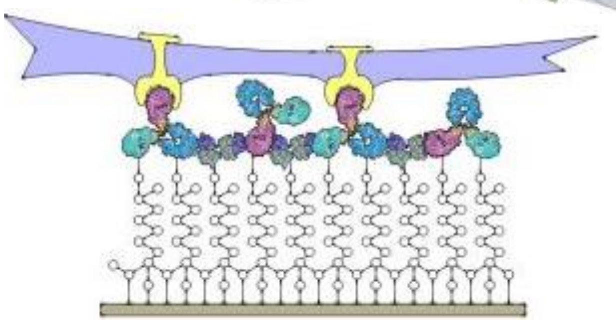
Ultrasensitive Acoustic transducer :

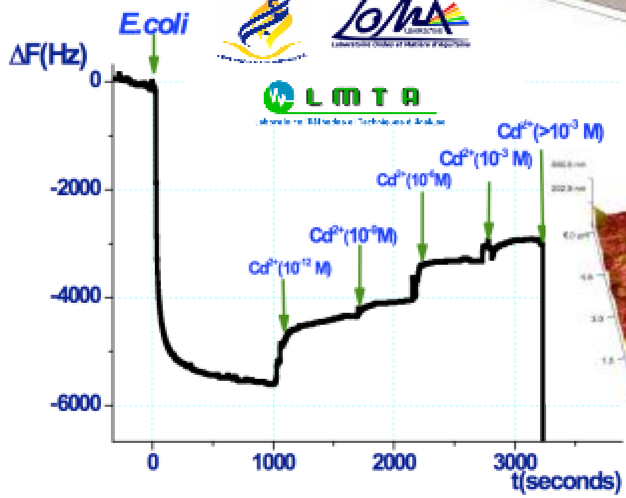
Application for bio sensing

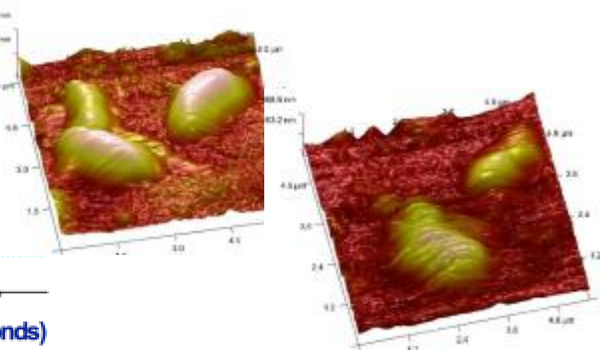
- Biological Species (bacteria, toxins) - Immunosensor
- Heavy metals with micro-organisms on multilayered polyelectrolyte film











Langmuir 09, Sens.Act.B 09-10-12-13, prix SBMicro 2010, ***PhD H.Tarbague & F.Fournel (2011)***
Bios.Bioelec 10-14, Sens.Act.B 14, ***PhD I.Gammoudi (2012)***

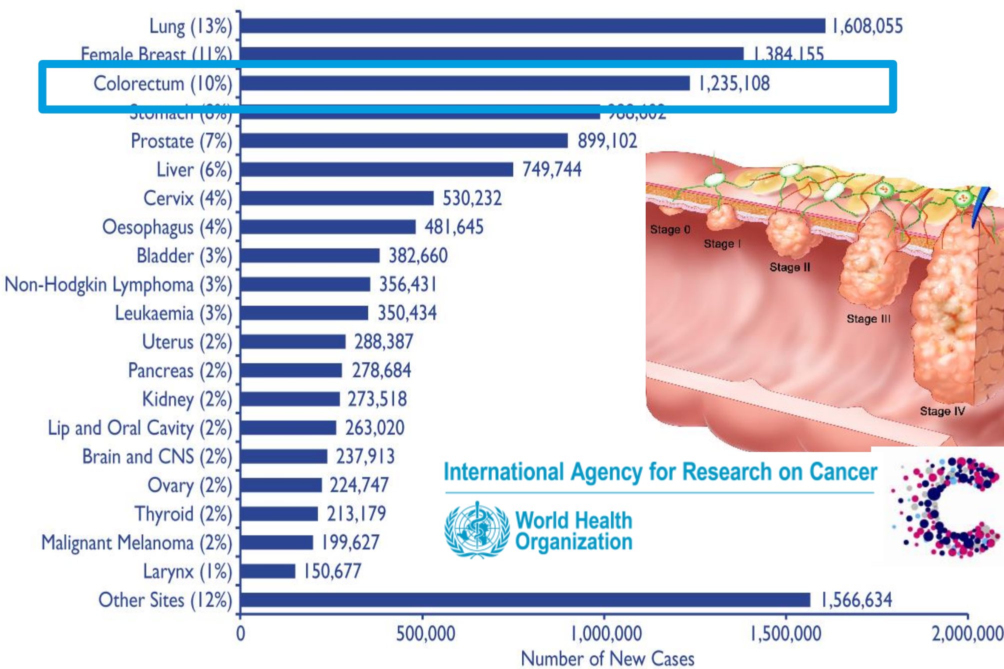
Dejous, C., Hallil, H., Raimbault, V., Rukkumani, R., & Yakhmi, J. V. (2016). Using Microsensors to Promote Development of Innovative Therapeutic Nanostructures.

Ultrasensitive Acoustic transducer :
Application for bio sensing

Application to cancer monitoring

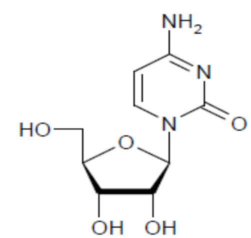
Projet ANR *CancerSensor*, CAPTEURS A EMPREINTES MOLECULAIRES POLYMERIQUES DE NUCLEOSIDES MODIFIES POUR UN SUIVI, NON-INVASIF, DE THERAPIES DE CANCERS

The 20 most commonly diagnosed Cancers worldwide

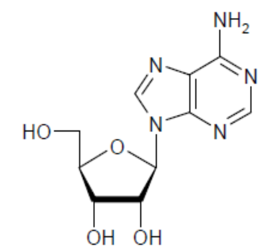


International Agency for Research on Cancer
World Health Organization
CANCER RESEARCH UK

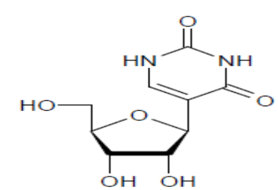
Colorectal Cancer biomarkers



Cytidine



Adenosine



Pseudouridine

Objective: Monitoring urinary nucleosides levels to evaluate therapies efficiency.

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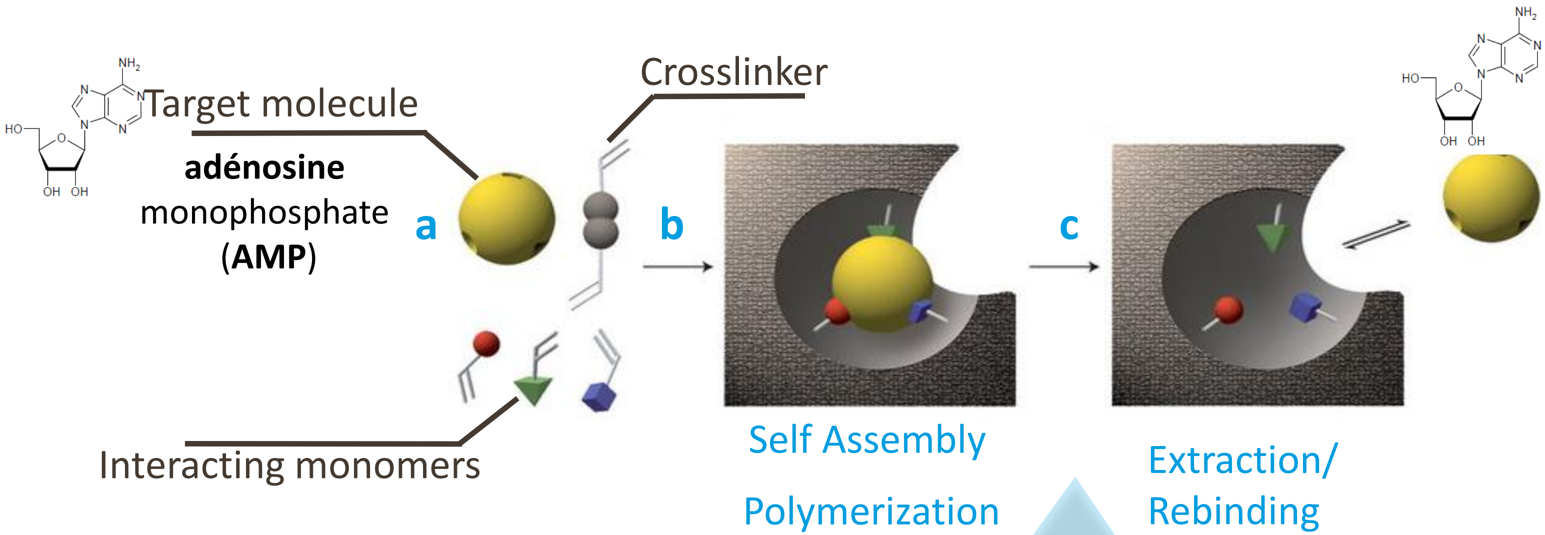
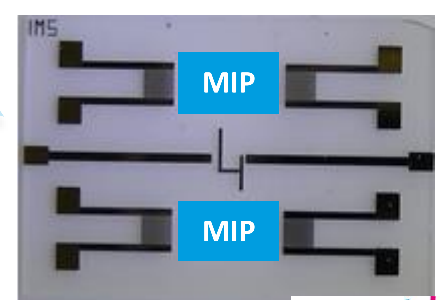
Ultrasensitive Acoustic transducer :

Application for bio sensing

Love-wave biosensor platform coated with thin film Molecularly Imprinted Polymers (MIPs)



- Inherent re-usability.
- Long-term stability and shelf life.
- Resistance to harsh environment.
- Low cost.

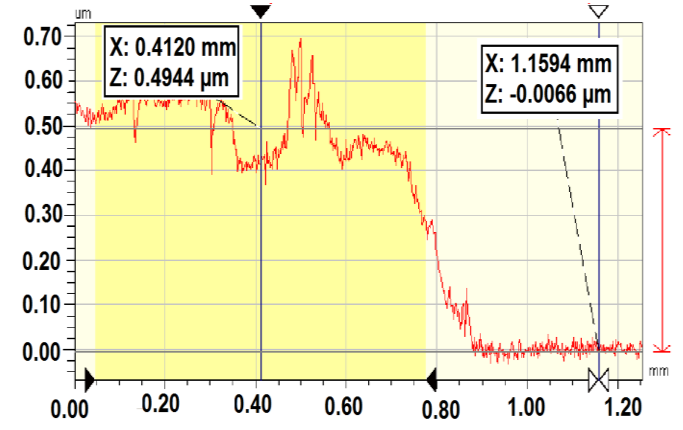


Ultrasensitive Acoustic transducer :

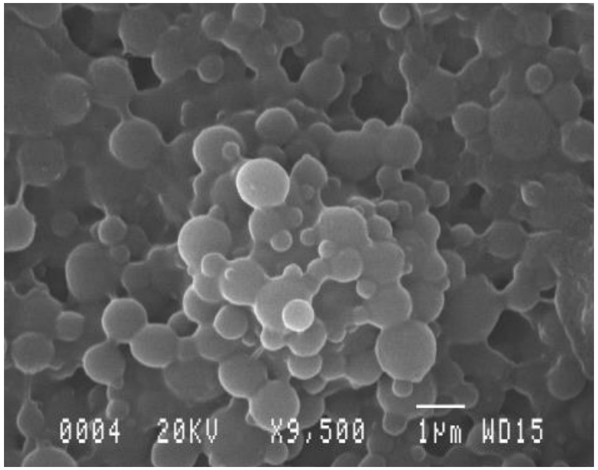
Application for bio sensing

Physical MIP film characterization

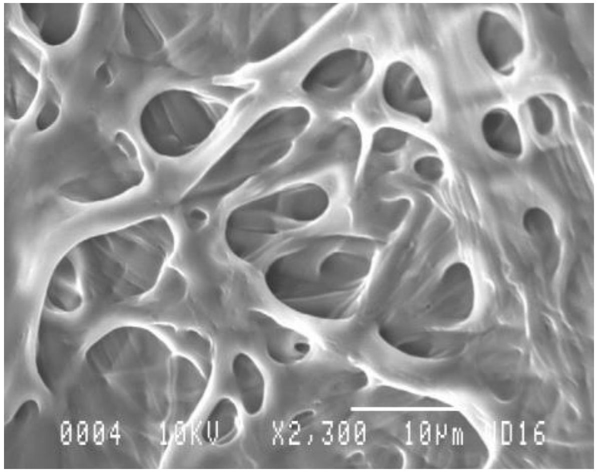
- Film thickness is about 500nm for both MIPs
- Homogeneity of the film is good for the MIP_{AMP} slightly less for the $MIP_{Pseudouridine}$
- Both films exhibits large specific areas



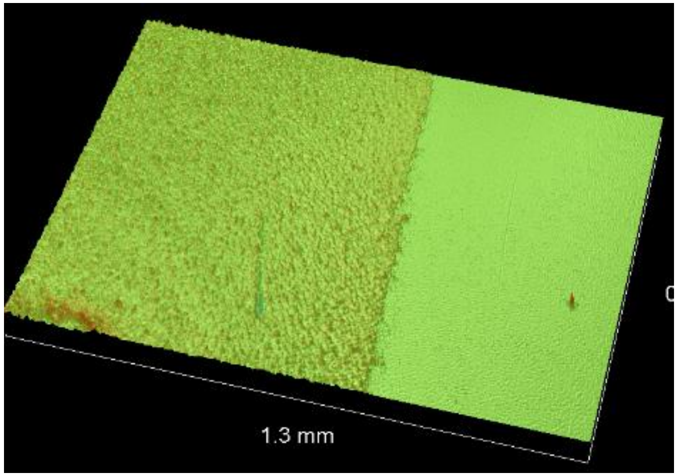
Stylus profilometer profile of a spin coated MIP_{AMP}.



SEM image of a spin coated MIP_{AMP} showing the porosity.



SEM image of a spin coated $MIP_{Pseudouridine}$ showing the porosity.



Optical profilometer view of a spin coated MIP_{AMP} showing the film uniformity.

N Lebal, H Hallil, C Dejous, B Plano, A Krstulja, R Delépée, L Agrofoglio, Dominique Rebière "Nucleosides analogues recognition by molecularly imprinted polymer-coated Love wave sensor" Micro & Nano Letters (2013) 8 (10), 563-566

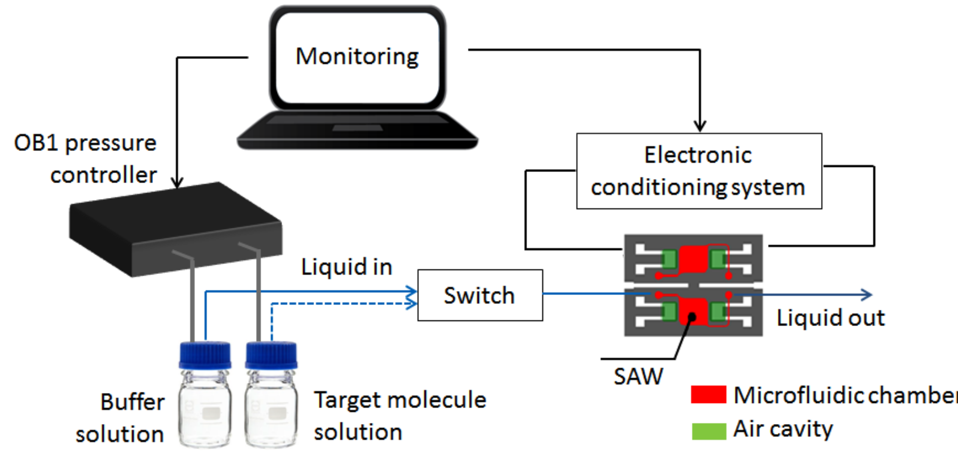
Ultrasensitive Acoustic transducer :

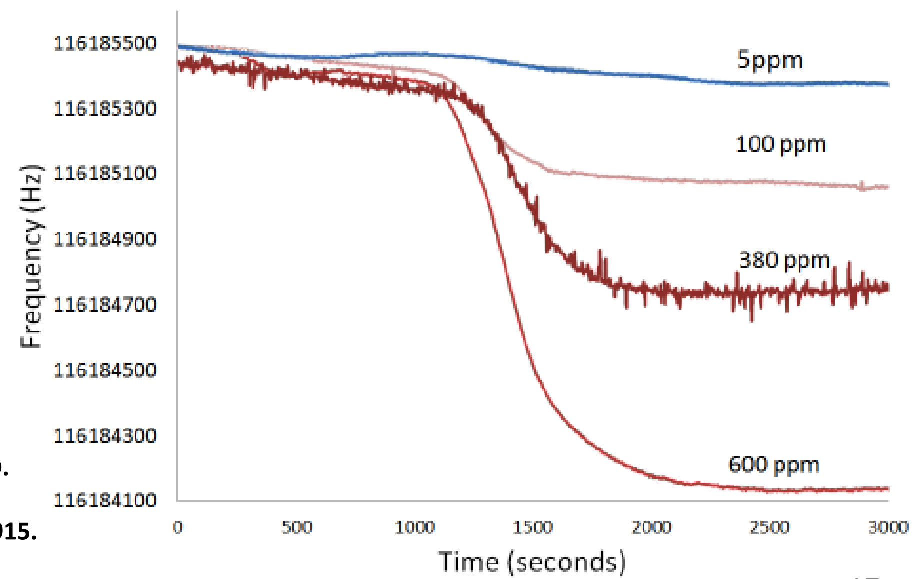
Application for bio sensing

MIP_{AMP}: Real-time nucleotide recapture experiment

- AMP template is extracted in bulk using a NH_3 / MeOH mixture
- Real-time recapture setup
- Injection of a buffer solution, followed by AMP solutions (5 to 600ppm)
- The oscillation frequency is monitored

✓ **150Hz shift is obtained for 5ppm of AMP.**

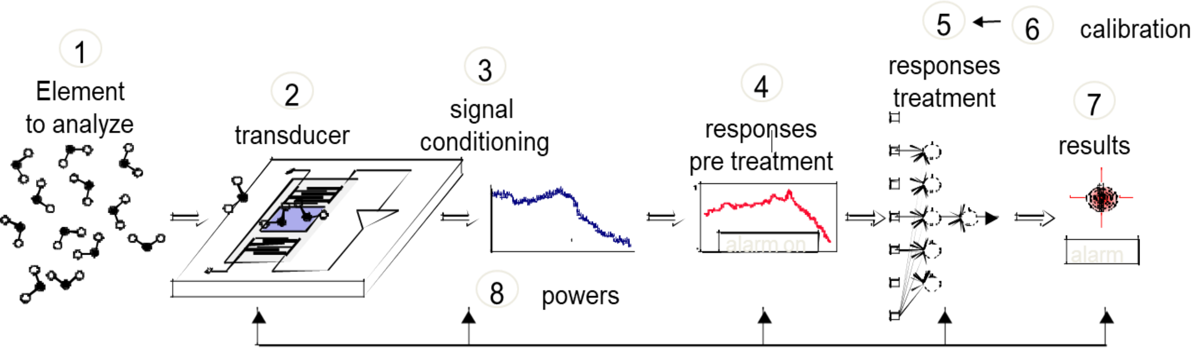




✓ **Feasibility of a Real-Time Diagnostic Tool for Colorectal Cancer**

Dejous, C., Hallil, H., Raimbault, V., Lachaud, J. L., Plano, B., Delépée, R., ... & Rebière, D. (2016). Love Acoustic Wave-Based Devices and Molecularly-Imprinted Polymers as Versatile Sensors for Electronic Nose or Tongue for Cancer Monitoring. *Sensors*, *16*(6), 915.

Les transducteurs à ondes acoustiques : des dispositifs adaptés aux mesures "tous terrains"



Très sensibles : ppt ou picogramme.

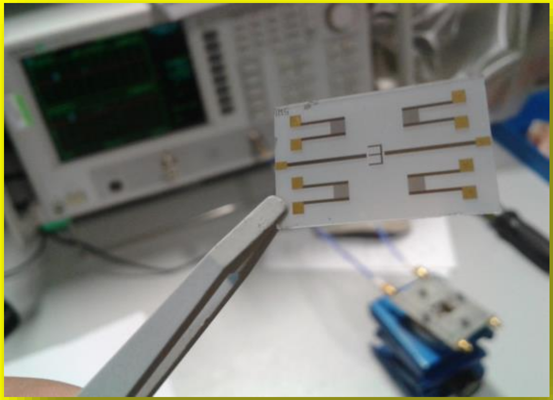
Détection en milieu gazeux et liquide

Portable, miniaturisable, intégrable

Temps de réponse rapide.

Pas de manipulation préalable de cibles

Laboratoire



Terrain

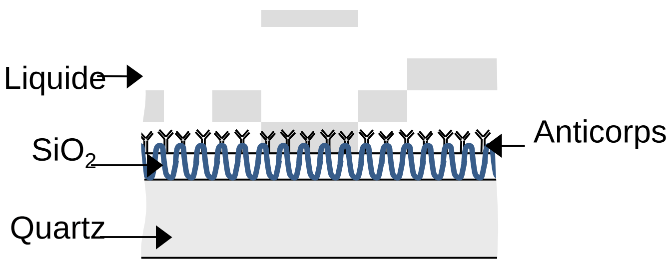
In situ

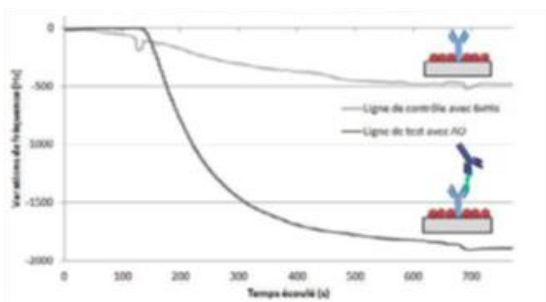
Vers des dispositifs plus sensibles, avec un électronique et un système de prélèvement évoluée pour une utilisation simple

Application à la détection de cibles de faible poids moléculaire (Cas des Cyanotoxines) ?

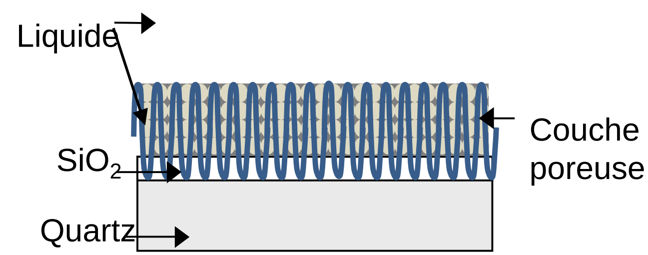
Solution proposée: couche mésoporeuse de TiO2 fonctionnalisée par anticorps monoclonaux anti-MCLR

fonctionnalisation de surface classique non appropriée

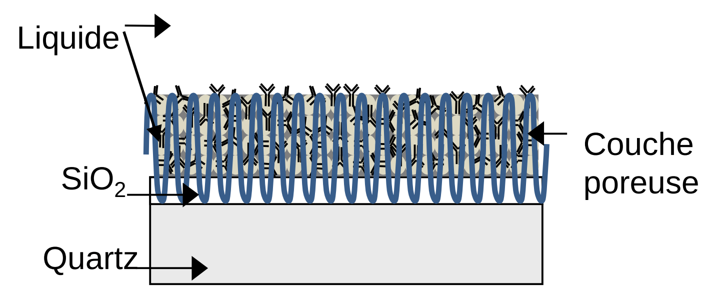




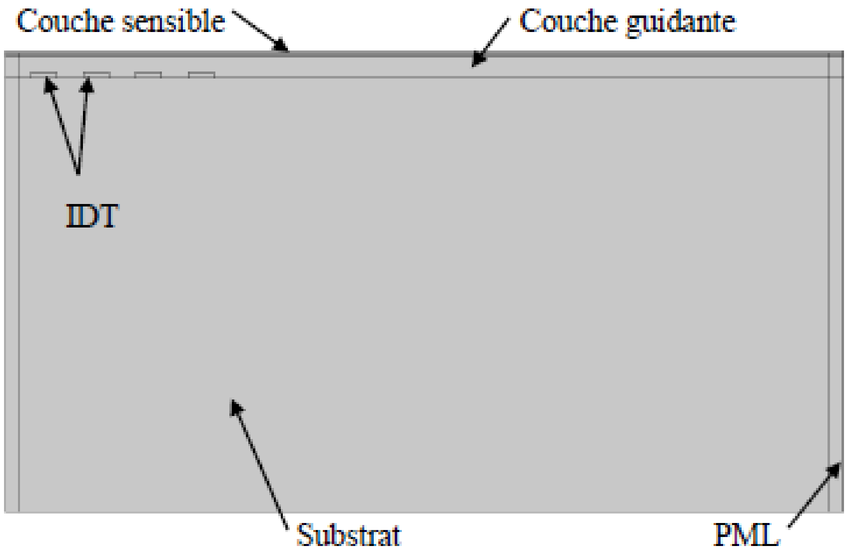
Confinement d'une partie de l'énergie acoustique dans le TIO2P => augmentation de la sensibilité gravimétrique du transducteur

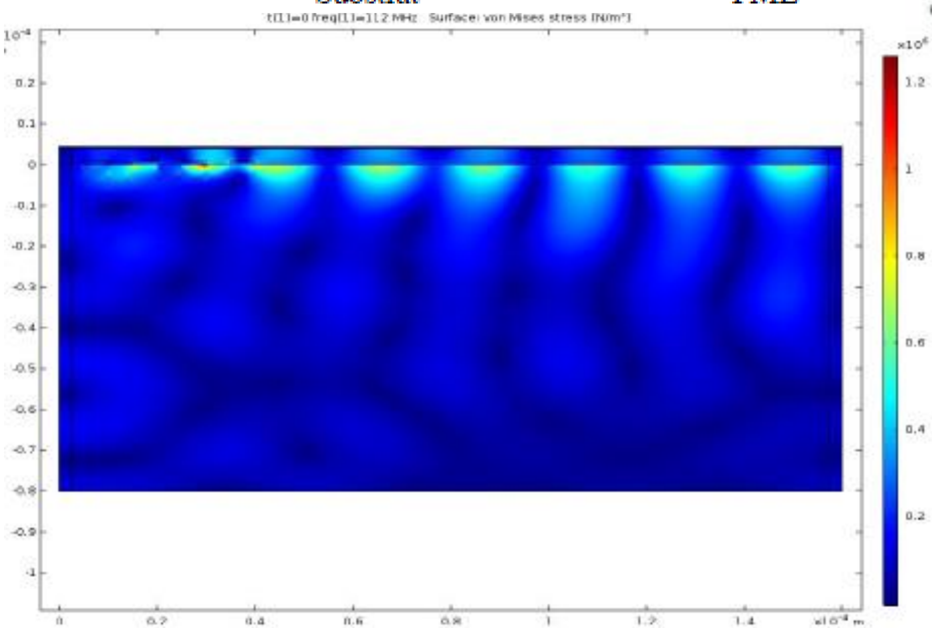


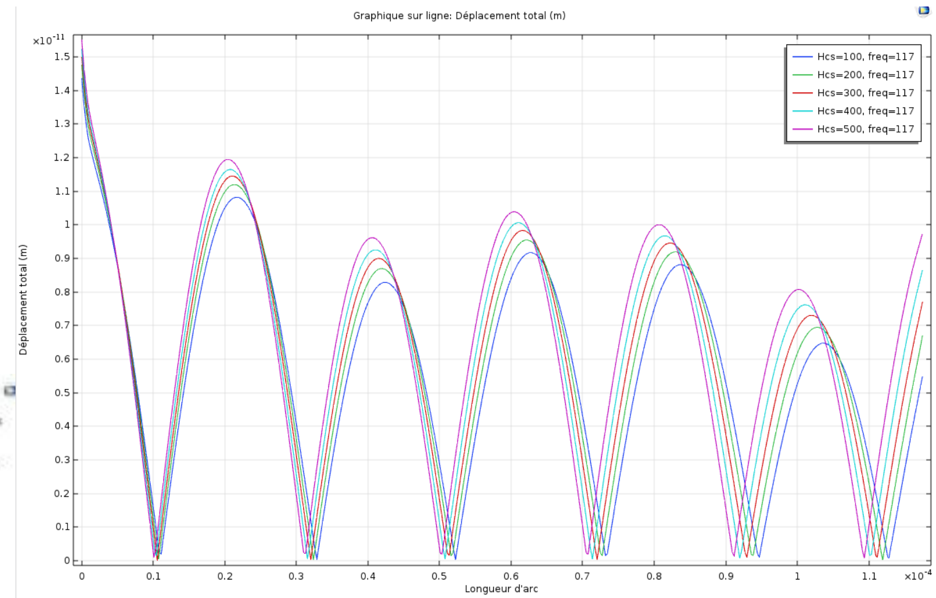
Augmentation de la surface spécifique => Plus d'anticorps « greffables » augmentation du potentiel de site de capture de cibles



Simulation par éléments finis : prise en compte des caractéristiques d'une couche poreuse en milieu liquide



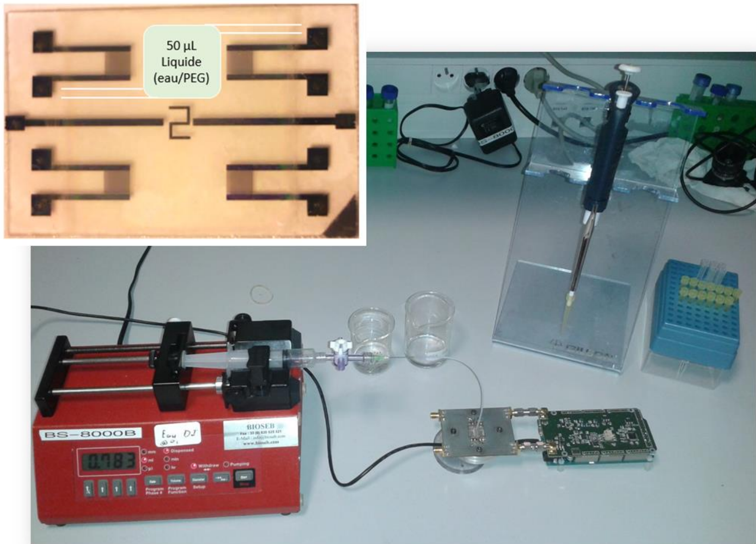


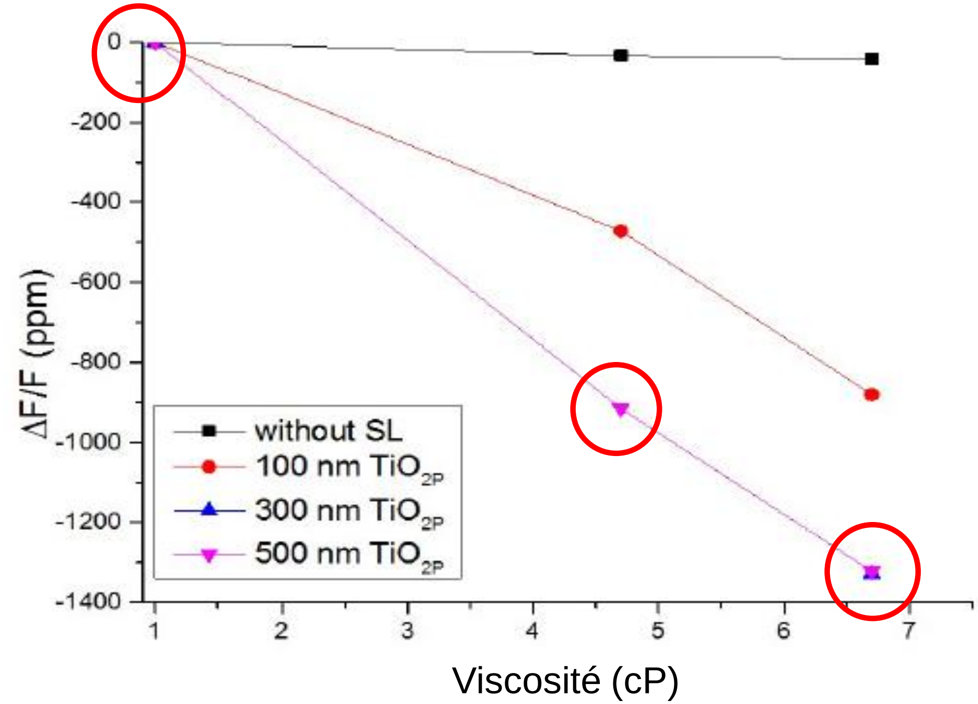


$\Rightarrow$ **Propagation d'une onde de Love possible avec une couche sensible de TIO_{2P}**

$\Rightarrow$ **Possibilité d'estimer l'énergie acoustique dans chacune des couches par simulation**

Sensibilité expérimentale du capteur à la viscosité





- ✓ Les dispositifs avec des couches sensibles de 300 nm et 500 nm deTiO2P sont **également plus sensibles** que le dispositif avec une couche sensible d'épaisseur 100 nm et 30 fois plus sensibles que l'appareil nu.

- ✓ Le fait que les sensibilités des dispositifs avec des couches sensibles d'épaisseurs 300nm et de 500 nm soient similaire peut être attribuée au **temps de diffusion** du liquide dans la couche poreuse de TiO2.

Conclusion

Capteur à transduction acoustique

- Micro-technologie maitrisée et totalement intégrable
- Plateforme générique: possibilité d'intégration des couches sensibles selon l'application visée
- Possibilité d'associer une puce microfluidique, ou des systèmes plus évolués pour des détections en milieu liquide (eau, urine, sang…etc)
- Très sensible et spécifique pour des applications biomédicales et santé environnementale (gaz toxiques, métaux lourds, bactéries, toxines, bio marqueurs nanoparticules fines,…etc)

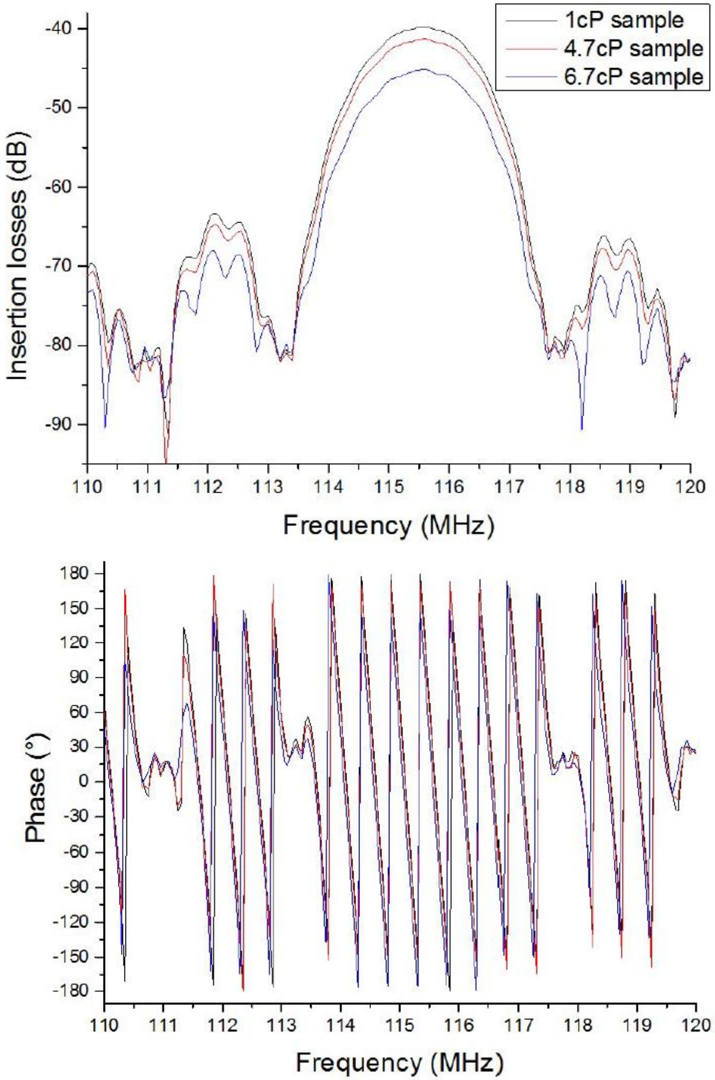
Capteurs tous terrains

- Transducteurs à ondes acoustique plus facilement « transportable » voire portable que les transductions actuellement utilisés pour des applications environnementales
- Possibilité de rendre le capteur « passif », et de désolidariser la tète de mesure du système d'interrogation électronique
- Interfaçage envisageable (R&D) sur des appareils du quotidien (PC – Smartphone)
- Faible cout de fabrication en production

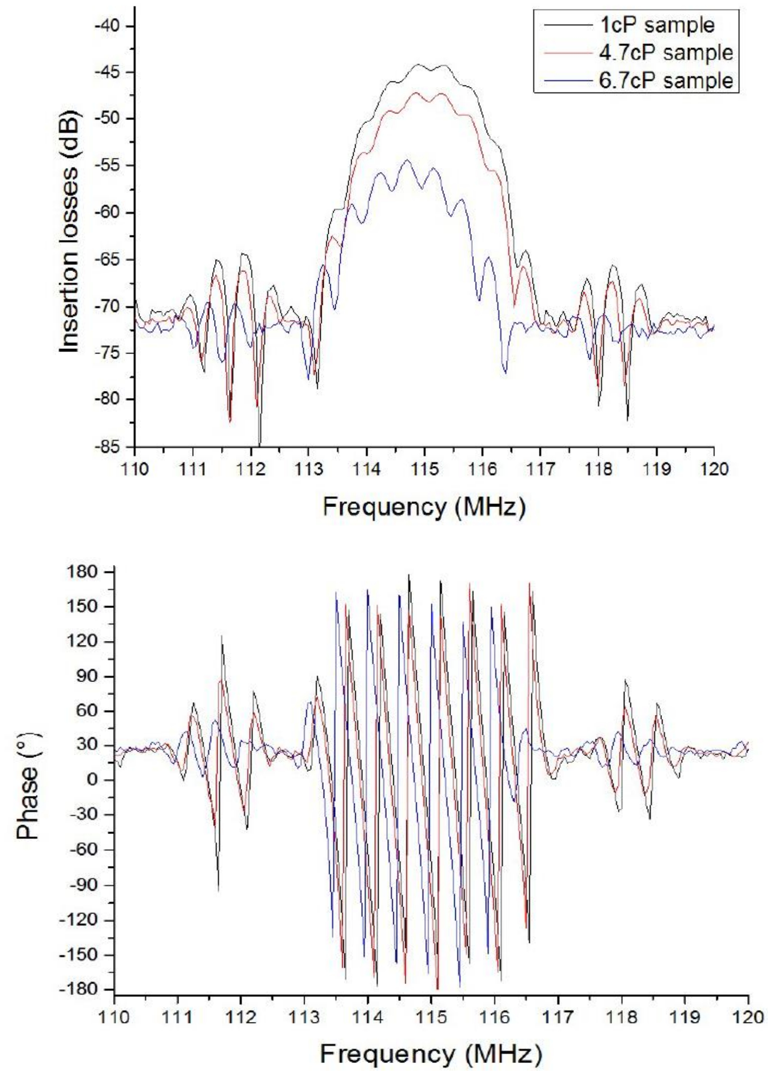
MERCI POUR VOTRE ATTENTION

Source : franceameriquelatine.org

Caractérisation électrique expérimentale : Interactions des liquides visqueux avec les ondes de Love



Capteur sans CS : interactions de surfaces



Capteur avec 300nm TiO2P : interactions de volume