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Graphene Oxide Inkjet-Printed Electronics: Electrochemical tuning of Charge Transport in Electrolyte-Gated Field-Effect Transistors and Biosensing Applications

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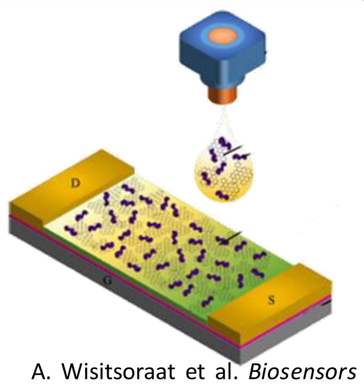
Introduction

Goal : use of functionalized inkjet-printed graphene oxide (GO) as an active material in an electrolyte-gated field effect transistor (EGFET) for biosensor applications.

A home-made graphene oxide surfactant-free printable ink (made of GO flakes) has been formulated and successfully printed by ink-jet on the photolithographed transistor structure. We have developed an *in-situ* electrochemical approach to obtain conductive reduced GO (rGO) directly on a bottom contact transistor architecture. We measured a non-linear effect of the reduction degree on charge transport properties in low voltage operating EGFETs. Moreover, our device has been used to monitor the metabolism of photosynthetic organisms.



A. Wisitsoraat et al. *Biosensors and Bioelectronics* (2017) 87,7

Graphene oxide processing for rGO-FET

1. Formulation of inkjet-printable graphene oxide ink

Ink content:

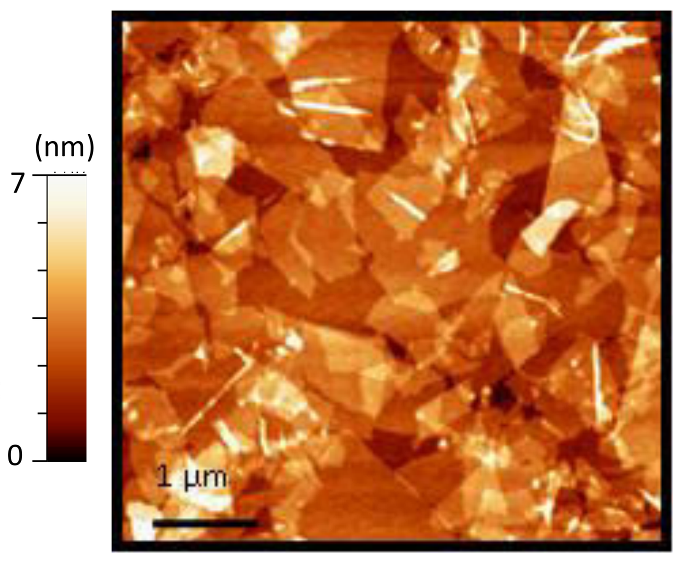
- GO (0.5 mg.mL^{-1}) in: 50% water, 20% ethylene glycol, 30% 1-propanol.

Ink rheological properties:

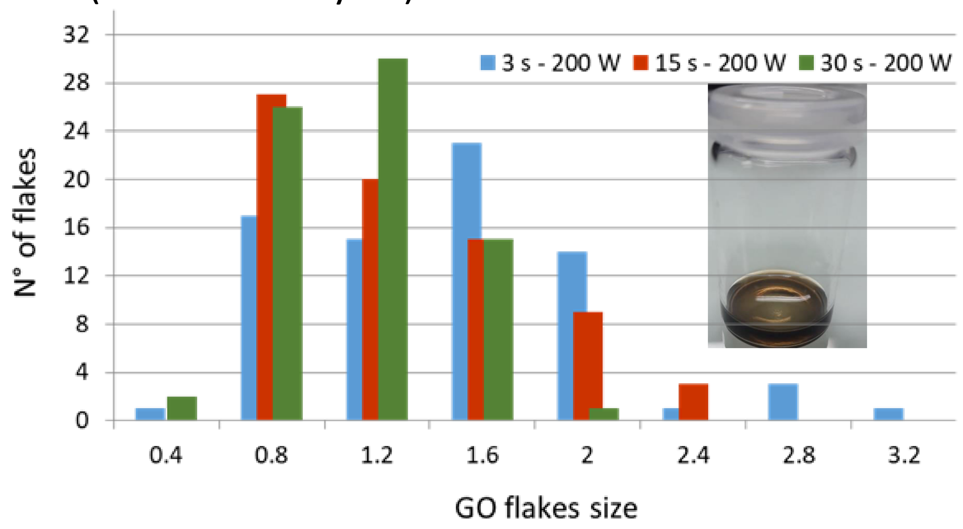
- surface tension: 28.3 mN.m^{-1}
- viscosity: 3.65 cP.

Control of GO flakes dimensions by ultrasonic fragmentation:

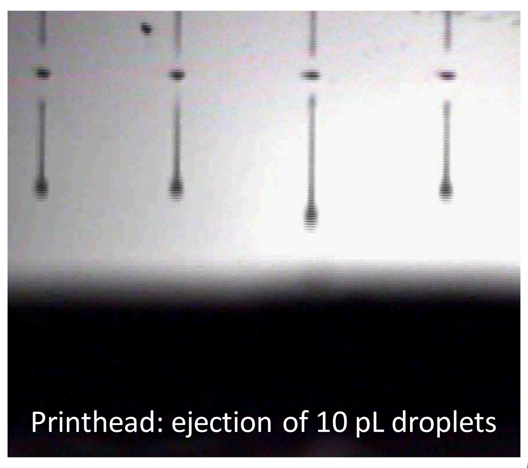
- lateral size 1 µm (prevents print head's nozzle clogging)
- average thickness 1-2 nm (2-6 atomic layers).



AFM image of GO flakes on SiO_2



Distribution of graphene flakes lateral size with fragmentation time



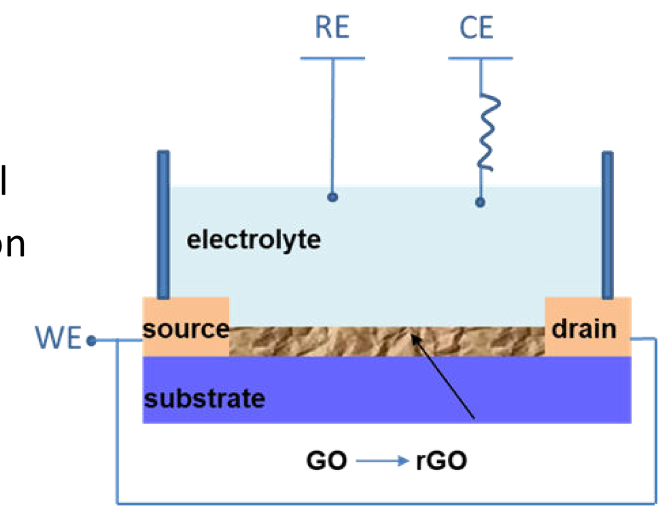
Printhead: ejection of 10 pL droplets

2. Electrochemical *in-situ* reduction

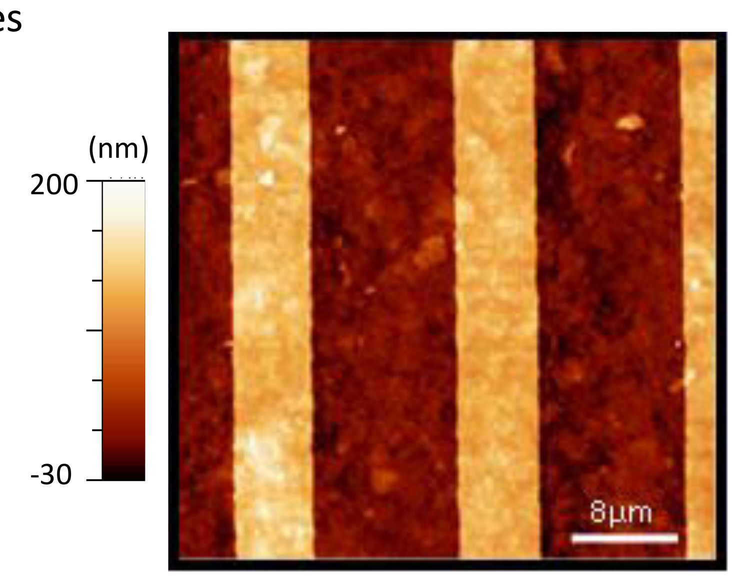
Reduction of GO printed film directly on GFET's electrodes (gold contacts made by photolithography).

Conditions:

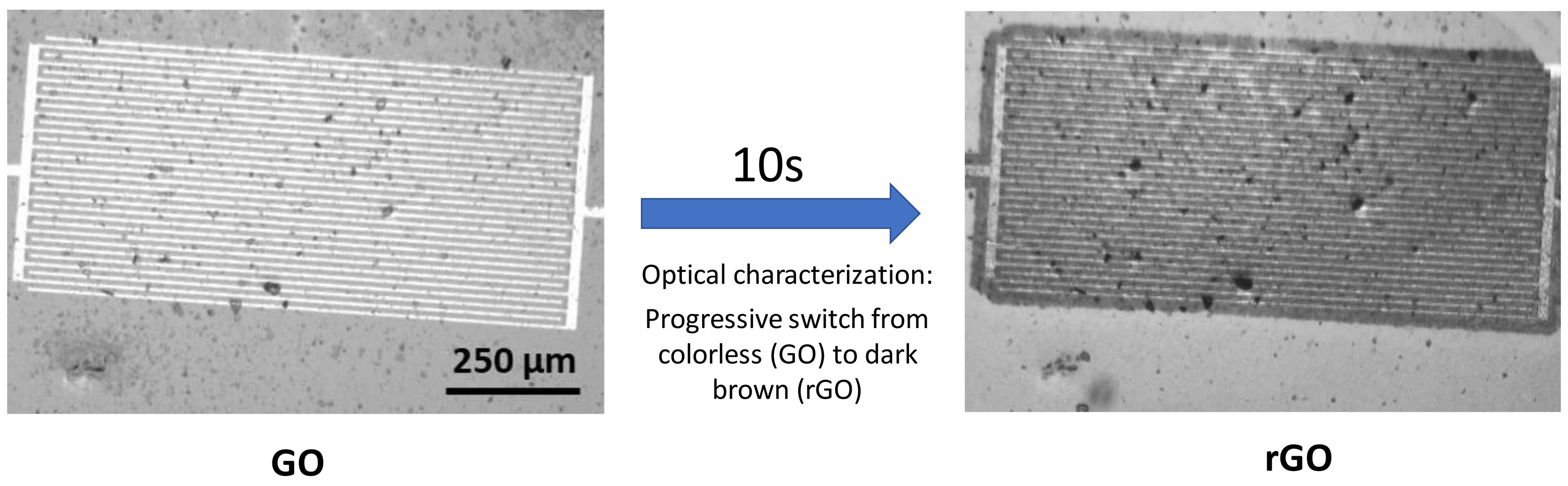
- potential: -1.3V vs Ag/AgCl
- various reduction time.



Electrochemical reduction setup



AFM image of inkjet-printed GO ink on bottom contact transistors' configuration

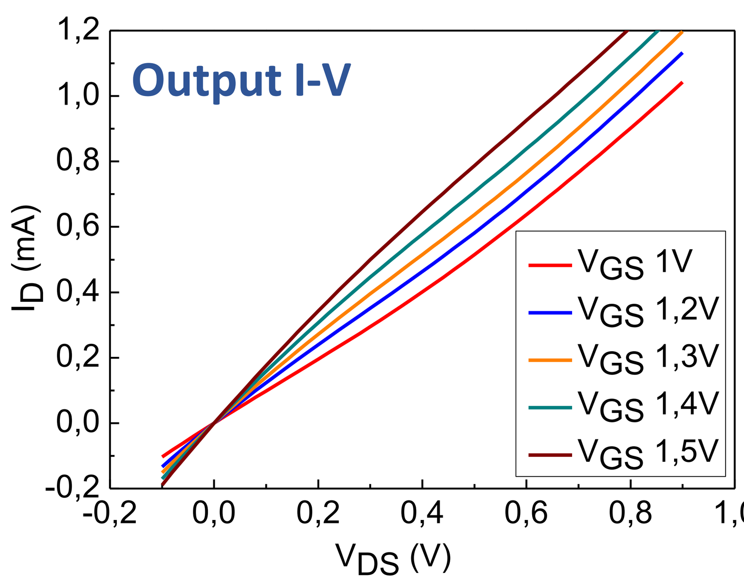


Optical characterization: Progressive switch from colorless (GO) to dark brown (rGO)

3. Electrical characteristics of rGO-FETs

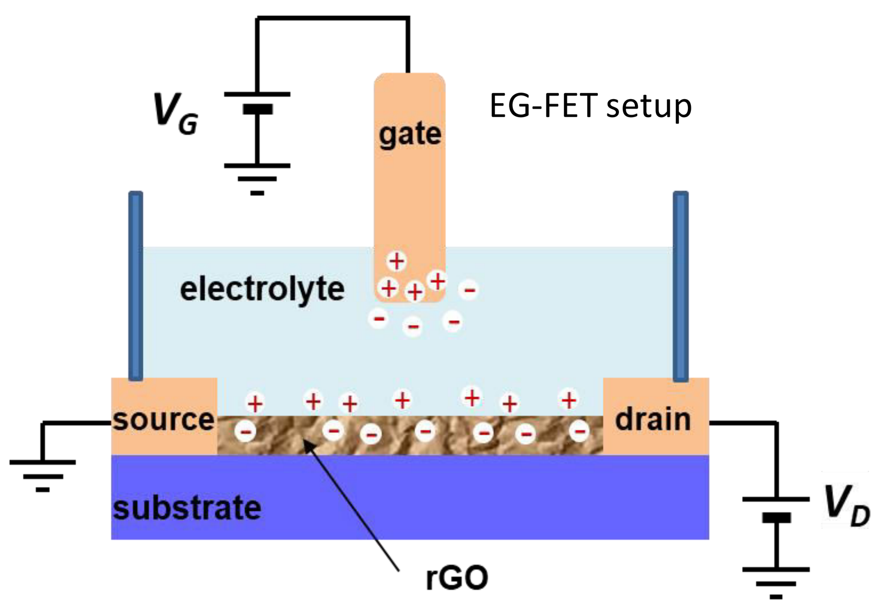
- Top-gate only (no back-gate)
- Electrolyte = phosphate buffer saline (PBS, 0.15 M NaCl).
- Modulation of charge carriers' concentration in rGO by applying appropriate gate voltage.
- The electrolyte-gated rGO-FET presents low operating voltages.
- rGO is characterized for different reduction yields.

Output I-V: field effect visible at low gate voltage.



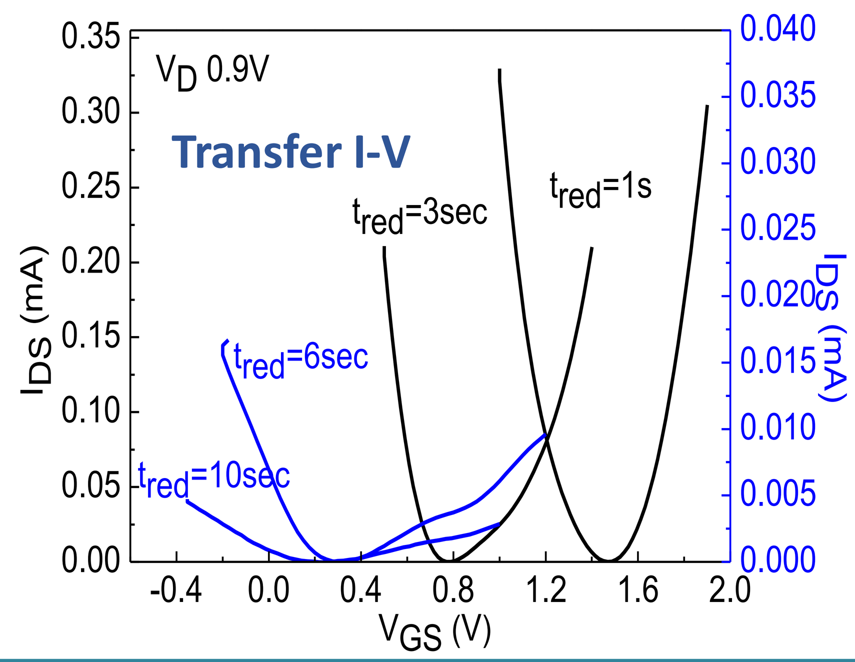
Reduction time effect: tuning of charge transport properties

- Estimated capacitance value 8 µF/cm².
- → the mobility strongly decreases versus t_{red}.



Transfer I-V: ambipolar behavior with a well-defined Dirac point (charge neutrality point).

- Shifting of Dirac point towards 0 with increasing reduction time (t_{red}) (may be due to dedoping effect expected upon GO reduction).
- Transconductance decreases upon reduction.

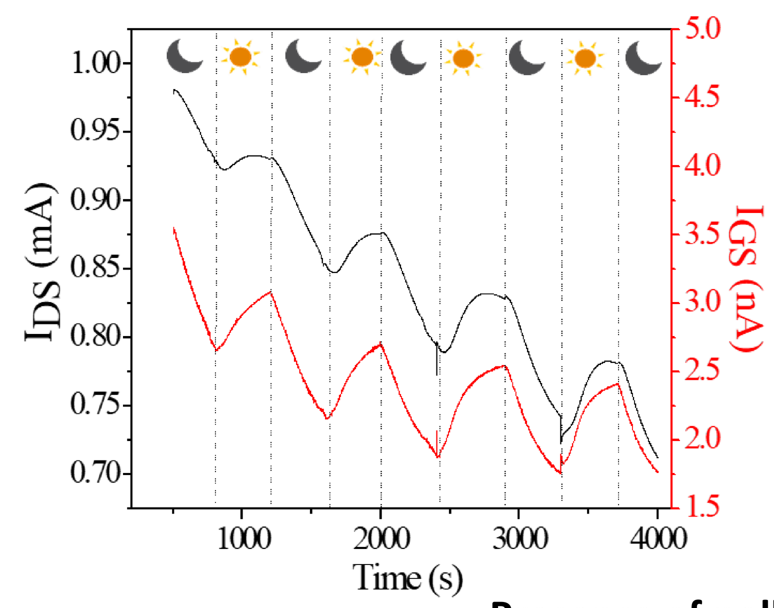


4. Biosensing

- rGO EG-FET has been employed for life-cycle monitoring of cyanobacteria's photosynthetic activity.
- Cyanobacteria (*Anabeana flosaquae*) used as an electrolyte.

☀ oxygen release in the presence of light

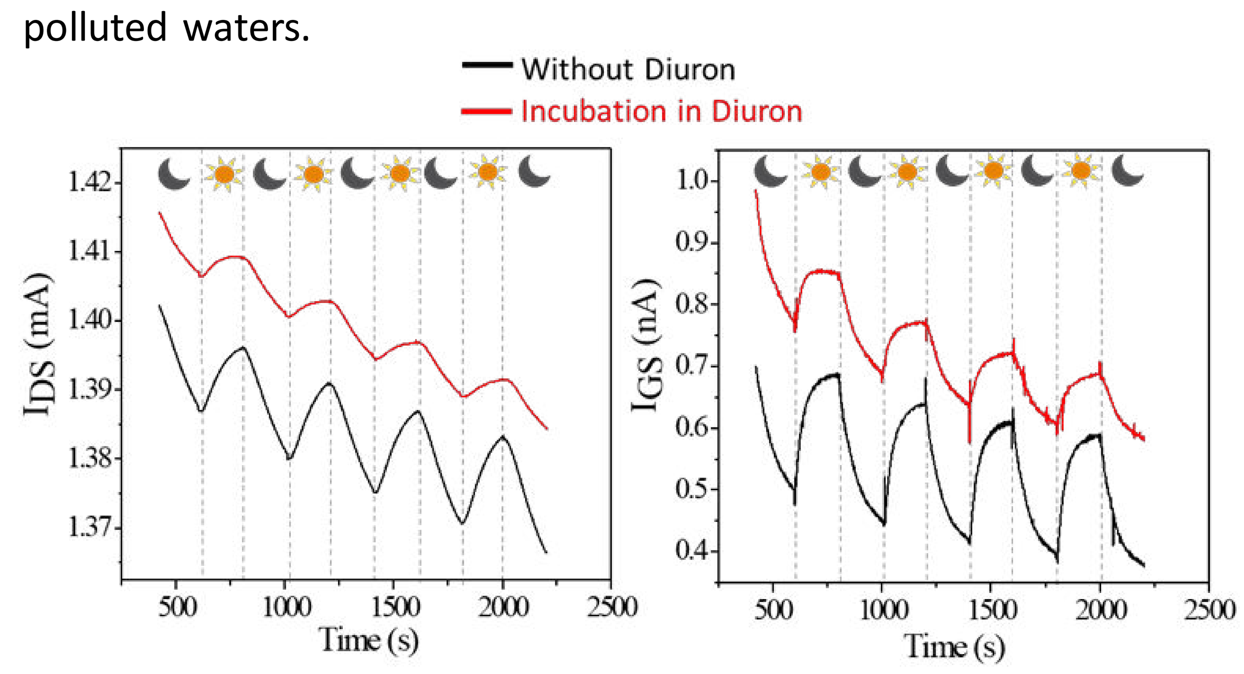
☾ oxygen decrease in the absence of light (breathing process)



- Both drain and gate current increase upon the device. illumination=oxygen concentration increase.
- Current decrease in the absence of light=oxygen concentration decrease.
- $\frac{I_{DS}}{I_{GS}} \approx 10^6$ amplification factor

Presence of pollutants

- Photosynthesis amplitude found to decrease upon the cyanobacteria incubation in herbicide solution (Diuron here used).
- Potential application of rGO EGGFET for qualitative prediction of polluted waters.



Conclusions

- ✓ Inkjet printing of lab-made GO ink.
- ✓ In-situ electrochemical reduction of GO into rGO on the transistor base.
- ✓ The reduction yield influences the electrical characteristics (I-V curves).
- ✓ Monitoring of algal photosynthetic activity and the pollutants presence.

Perspectives

- ➢ Fine tuning of doping effects in rGO through electrochemical and molecular approach.
- ➢ Statistical study of different herbicide influence on the algal metabolism.

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