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Electrochemical Characterization of Graphene-based Transducer for Biosensor Development

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ABSTRACT

Electrochemical characterization of various deposition methods of reduced graphene oxide (rGO) on glassy carbon electrode (GCE) was studied. Parameters that were varied include graphene oxide (GO) concentration, GO drying time, number of GO electrochemical reduction cycle, and amount of added gold nanoparticles (AuNPs) in enhancing electrical conductivity of transducer layer. The reduced graphene oxide gold nanoparticles (rGO-AuNPs) composite transducer layer was fabricated via a simple two-step physical (drop-cast) and subsequent electrochemical reduction. Cyclic voltammetry (CV) was used to characterize redox capability of the transducer layer. Electrochemical deposition of GO suspension with concentration of 6.2 mg/ml gives higher anodic peak current, I_{pa} (+10.061 mA) when compared to most peak current reported in literature for a 3 mm inner diameter electrode; therefore is an excellent precursor for the development of redox active transducer that results in highly sensitive biosensors.

INTRODUCTION

Graphene is a monolayer sheet of sp²- bonded carbon atoms arranged in perfect hexagonal lattice

Chemical	Electronics	Structural	Thermal
High surface area to volume ratio (2,630 m²/g)	High electron mobility; High electrical conductivity	High strength (300x steel); High rigidity (tensile strength 1 TPa)	Excellent thermal conductivity

Perfect as transducer!

Commercial potentials:

- ❑ Low-cost production of a highly-sensitive biosensor
- ❑ Continuous improvement on the overall performance of a biosensor

Novelties:

- ❑ Use of ultra highly concentrated GO (UHC GO) that is commercially available
- ❑ Oxidation peak current, I_{pa} obtained is significantly higher than those reported in literature
- ❑ Rubbing on GO sheet to deposit graphene on GCE

METHODOLOGY

Objectives:

○ To achieve highest oxidation peak current, I_{pa} after deposition of rGO on GCE to enhance biosensor sensitivity

○ To electrochemically characterize transducer layer using cyclic voltammetry via 3-point electrode setup and a potentiostat/galvanostat

○ To characterize the effects of two graphene precursors:

1) UHC GO suspension: Ultra highly concentrated single-layer graphene oxide (concentration of 6.2 mg/ml)
2) GO sheets

Deposition of graphene-based composite on glassy carbon electrode (GCE)

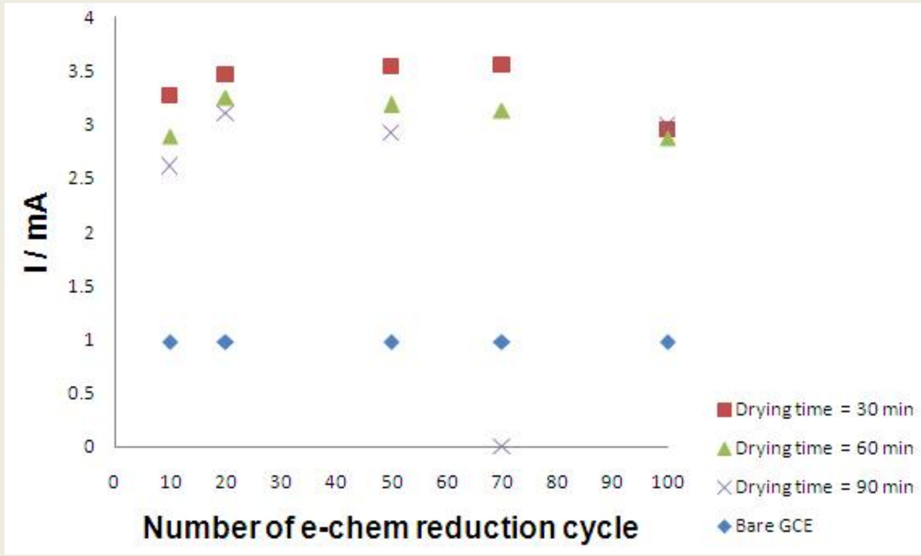
- **Physical**
  - Dip-coat in **UHC GO** suspension & reduce
  - Drop-cast **UHC GO** suspension & reduce
    - Drop-cast **UHC GO + AuNPs** composite & reduce (800:1 ratio)
  - Rub on **GO sheet** & reduce
- **Electrochemical**
  - Electrodeposit **UHC GO** suspension & reduce
    - Electrodeposit **UHC GO + AuNPs** composite & reduce

RESULTS & DISCUSSION

○ Dip-coat method (UHC GO)

Dip-coat time (min)	Drying time (min)	Anodic peak current, I_{pa} (mA)						
		Bare GCE	GCE/GO	GCE/rGO (10 cycle)	GCE/rGO (20 cycle)	GCE/rGO (50 cycle)	GCE/rGO (70 cycle)	GCE/rGO (100 cycle)
15	30	0.978	0.0023	3.267	3.462	3.538	3.559	2.951
15	60		0.0016	2.891	3.253	3.188	3.137	2.874
15	90		0.0015	2.614	3.101	2.923	-	2.996

Highest $\triangle I_{pa}$ **+2.581 mA**



Dip-coating is a practical and easy deposition method on electrodes surface.

Shorter drying time with optimum number of electrochemical reduction between 20 to 70 cycles results in highest peak current.

Fig. 1 Current vs. reduction cycles of GCE/rGO in 100 mM $Fe(CN)_6^{3-/4-}$ solution at scan rate of 100 mVs^{-1} for different drying time for electrodes modified via dip-coating.

○ Drop-cast method (UHC GO & UHC GO-AuNPs)

Nanomaterial	Sample Size	Cost per sample	Highest ∆ Ipa
UHC GO	8 µL	RM0.0400	+4.329 mA
UHC GO + AuNPs	8 + 2.5 µL	RM0.0475	+5.988 mA

30 minutes drying in ambient air after drop-casting and 20 electrochemical reduction cycle to get highest $\triangle I_{pa}$

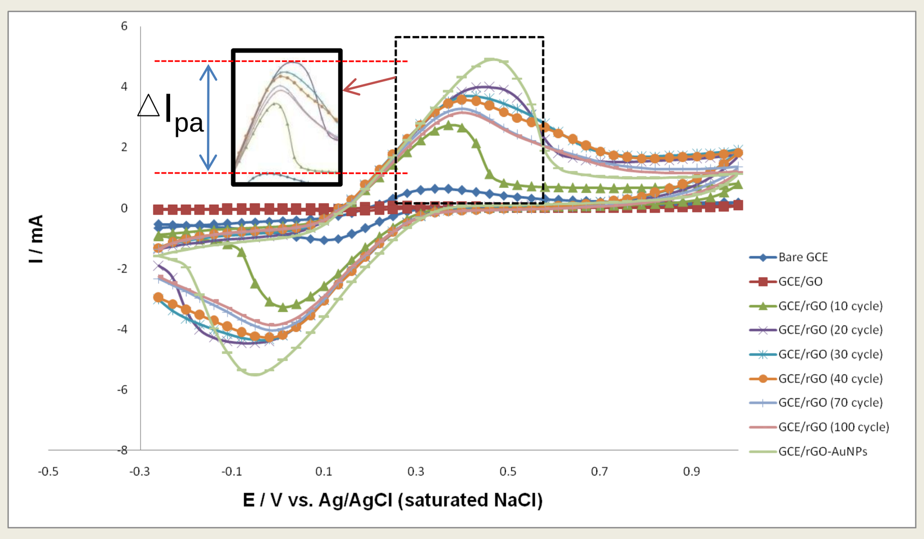


Fig. 2 CVs of GCE/rGO and GCE/rGO-AuNPs in 100 mM $Fe(CN)_6^{3-/4-}$ solution at scan rate of 100 mVs^{-1} for drop-cast deposition method

○ Rubbing method (GO sheet)

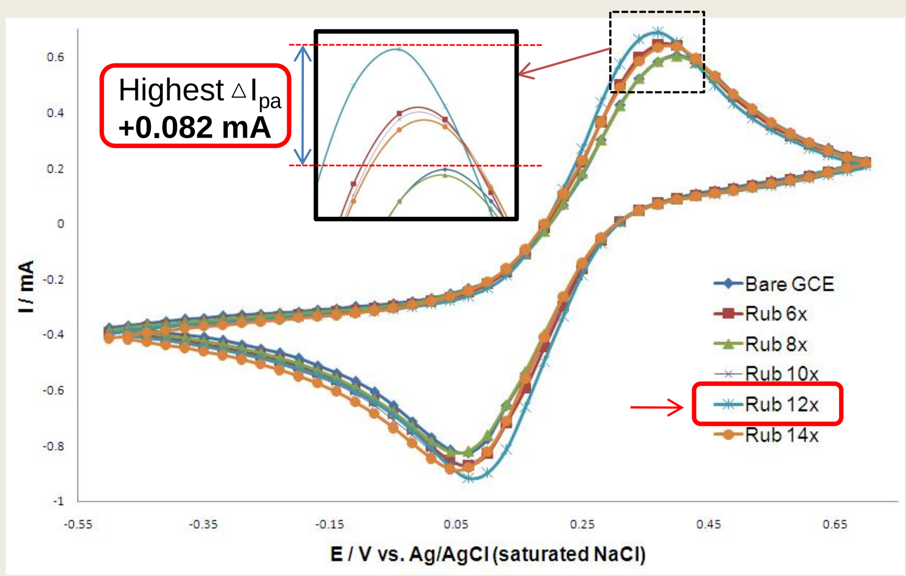


Fig. 3 CVs of GCE/rGO in 100 mM $Fe(CN)_6^{3-/4-}$ solution at scan rate of 100 mVs^{-1} for deposition method via rubbing

Fixed variables:

- ○ GO sheet thickness
- ○ Direction of rubbing
- ○ Side of GO sheet

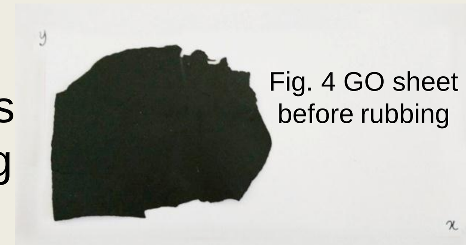


Fig. 4 GO sheet before rubbing

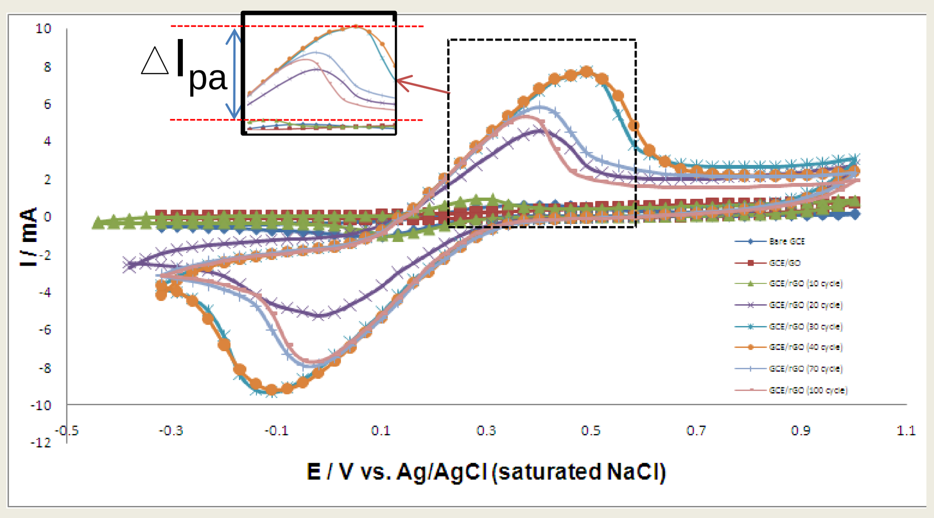
New method to deposit graphene on GCE

For ±1mm GO sheet thickness, optimum # of rubbing is 12x

Highest $\triangle I_{pa}$ measured comparable to that reported in current literature

○ Electrochemical deposition method (UHC GO)

Highest $\triangle I_{pa}$ +10.091 mA!



One electrodeposition cycle + 15 min dry time

30 to 40 electrochemical reduction cycle to get highest $\triangle I_{pa}$

Fig. 5 CVs of GCE/rGO in 100 mM $Fe(CN)_6^{3-/4-}$ solution at scan rate of 100 mVs^{-1} for electrochemical deposition method

○ SUMMARY

Deposition method	Dip-coat	Drop-cast ★	Rubbing	E-chem ★
Highest peak current, I_{pa} (GCE/rGO)	+2.581mA	+4.329mA	+0.082mA	+10.061mA
Highest peak current, I_{pa} (GCE/rGO-AuNPs)	-	+5.988mA	-	On-going

CONCLUSIONS

1. UHC GO suspension with concentration of 6.2 mg/ml gives higher anodic peak current, $\triangle I_{pa}$ (+10.061 mA) when compared to most peak current reported in literature; therefore is an excellent precursor for the development of redox active transducer layer and subsequently highly sensitive biosensors.
2. Cost of UHC GO and AuNPs is only ~RM0.04 per 8 µL and RM0.0075 per 2.5 µL, respectively, hence, the materials are attractive for developing low-cost biosensors.
3. GO sheets can be used as a precursor for graphene transducer and deposition on GCE can be done via rubbing method on the GO sheets.

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