

Supporting Information

Charge Transfer Driven by Redox Dye Molecules on Graphene Nanosheets for Room- Temperature Gas Sensing

Wenbo Liu^{1,2}, Junwei Zeng^{1,2}, Yixun Gao^{1,2}, Hao Li^{1,2}, Nicolaas Frans de Rooij², Ahmad Umar³, Hamed Algarni⁴, Yao Wang^{1,2,} and Guofu Zhou^{1,2}*

¹Guangdong Provincial Key Laboratory of Optical Information Materials and Technology, Institute of Electronic Paper Displays, South China Academy of Advanced Optoelectronics, South China Normal University, Guangzhou, 510006, P. R. China.

²National Center for International Research on Green Optoelectronics, South China Normal University, Guangzhou, 510006, P. R. China.

³Promising Centre for Sensors and Electronic Devices, Department of Chemistry, Faculty of Science and Arts, Najran University, Najran, 11001, Kingdom of Saudi Arabia.

⁴Department of Physics, King Khalid University, Abha, 61421, Kingdom of Saudi Arabia.

* *Corresponding Author: Yao Wang, Email: wangyao@m.scnu.edu.cn*

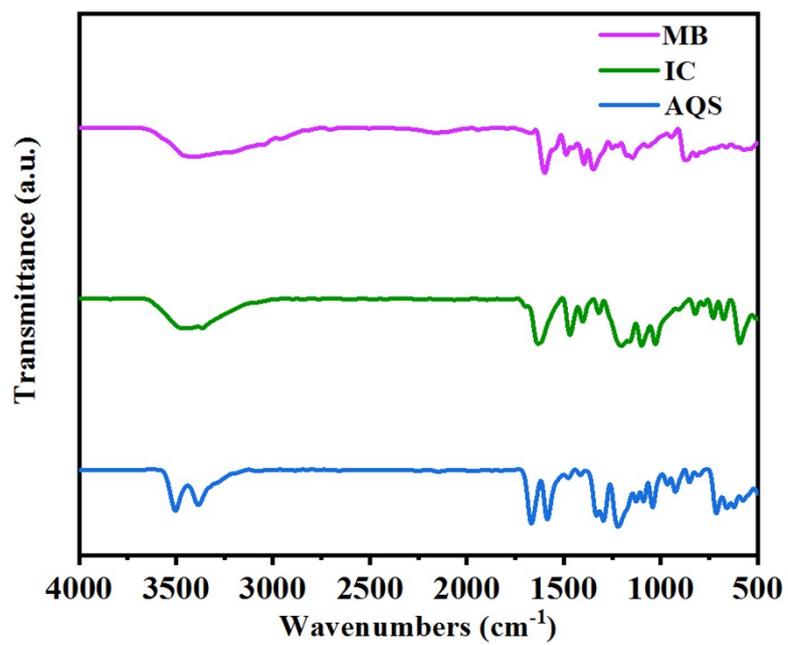


Figure S1. FTIR spectra of MB, AQS and IC.

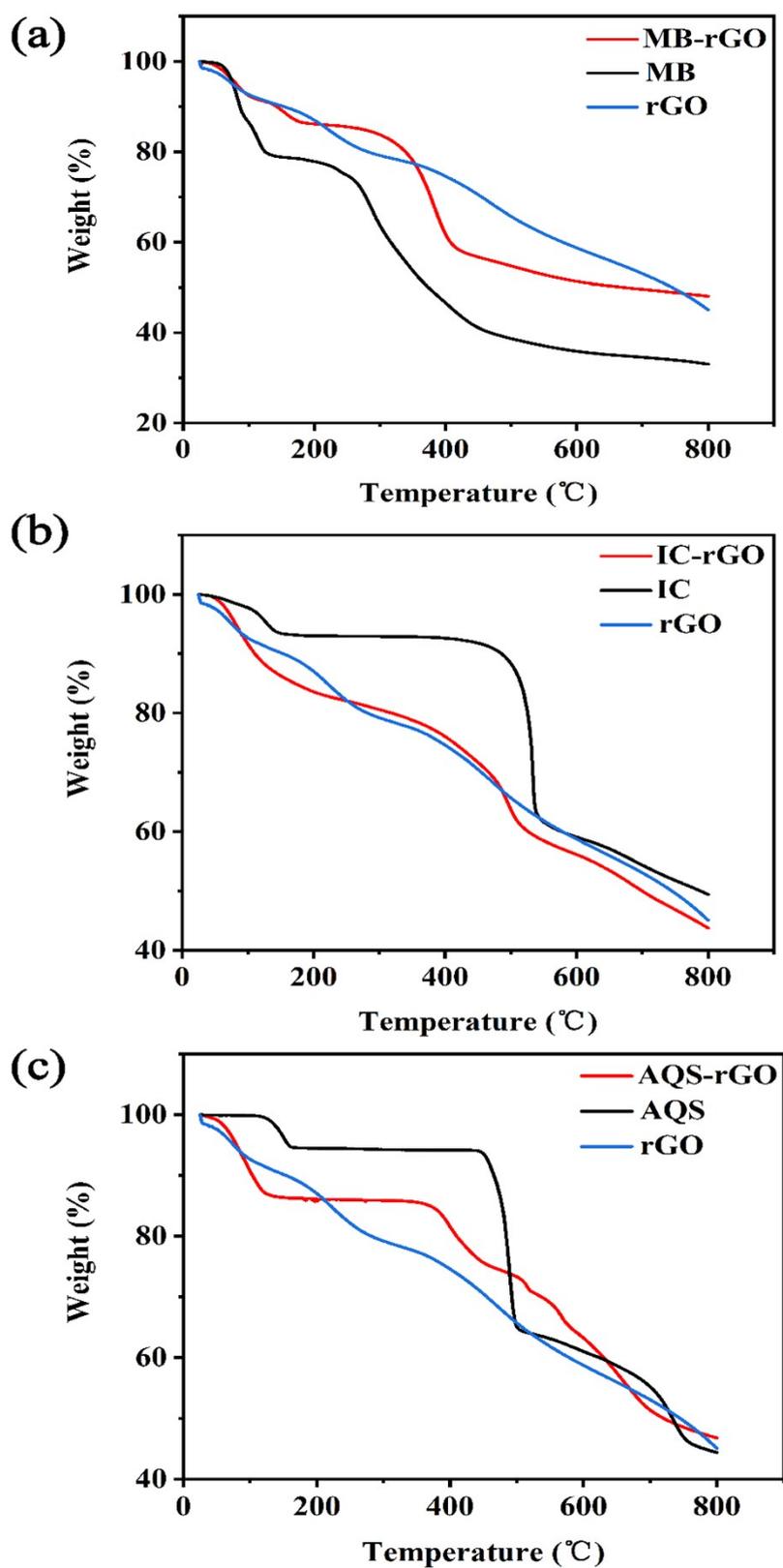


Figure S2. TGA curves of (a) rGO&MB&MB- rGO, (b) rGO&IC&IC- rGO, and (c) rGO&AQS&AQS-rGO composite with different mass ratios from room temperature to 800 °C under N₂ atmosphere.

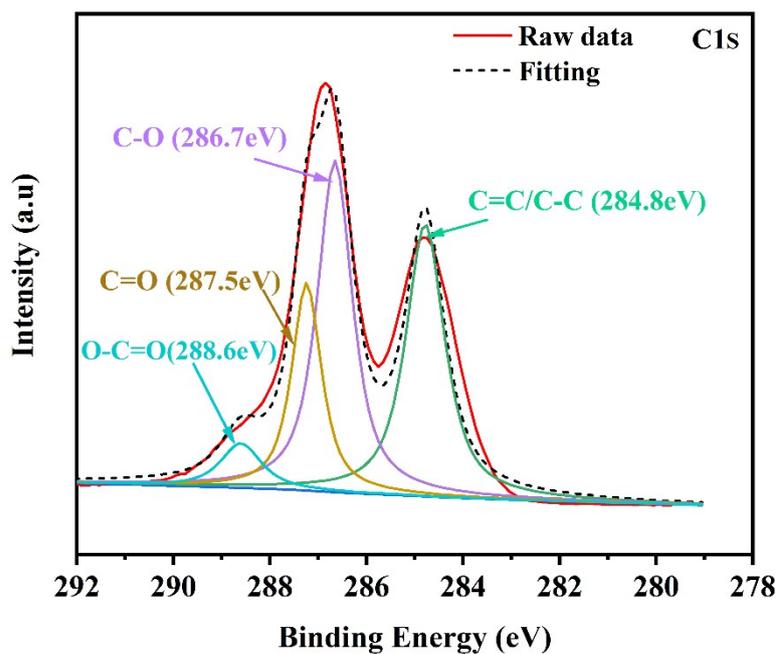


Figure S3. XPS survey spectrum C 1s of GO.

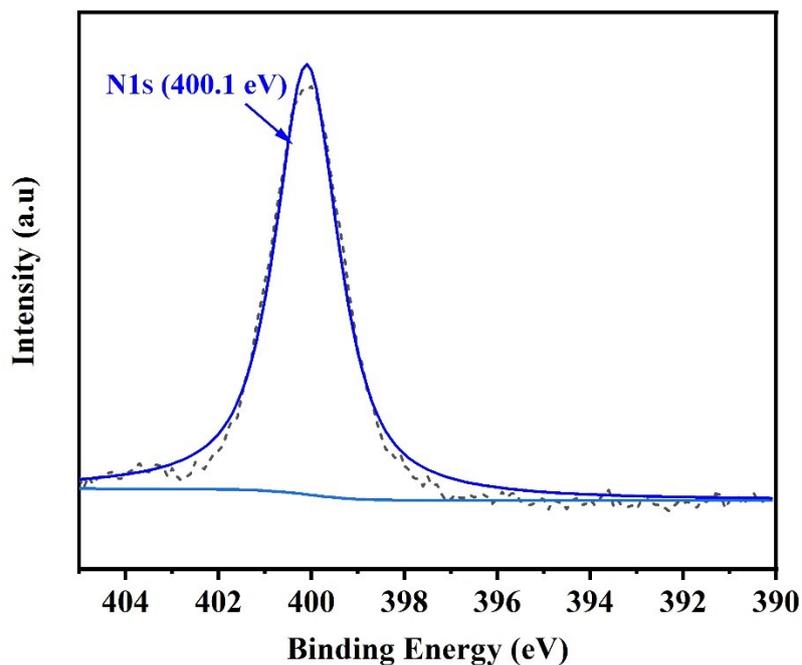


Figure S4. XPS survey spectrum N 1s of IC-rGO.

The appearance of the characteristic peak N 1s at 400.1 eV in the XPS spectrum confirms the successful complexation of IC molecules with rGO via π - π stacking (Figure S4).

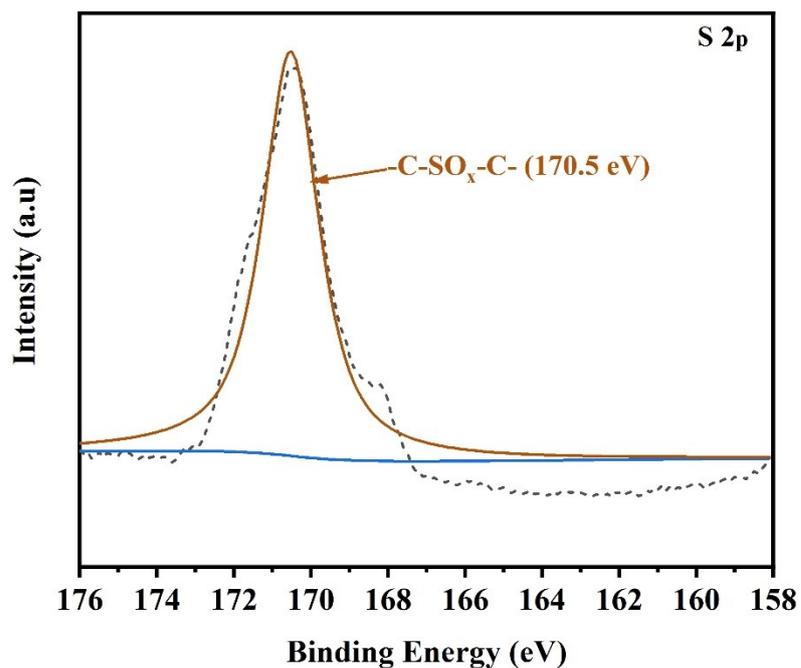


Figure S5. XPS survey scan spectrum S 2p of AQS-rGO.

Figure S5 shows that the peak position of the S 2p spectrum at 170.5 eV is attributed to the sulfonate functional group (-C-SO_x-C-),¹ indicating the successful complexation of AQS with rGO.

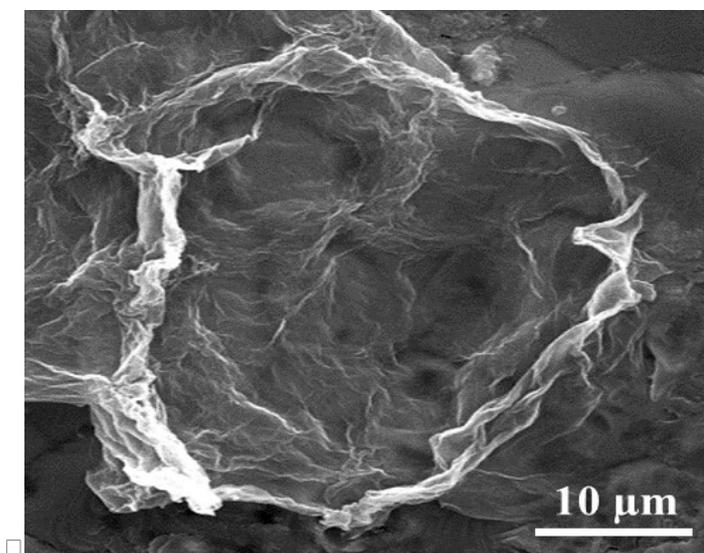


Figure S6. SEM images of rGO deposited on the surface of IDEs.

The gas sensors are made by the drip-drying method (Figure S7a). Using Keithley 2450 source meter to record changes in sensors' resistance before and after NO₂ exposure, and the response is calculated by the ratio of resistance captured in an atmosphere of air (R_a) and NO₂ (R_g) respectively, i.e. $S=R_a/R_g$. The schematic illustration of gas sensing tests is shown in Figure S7b.

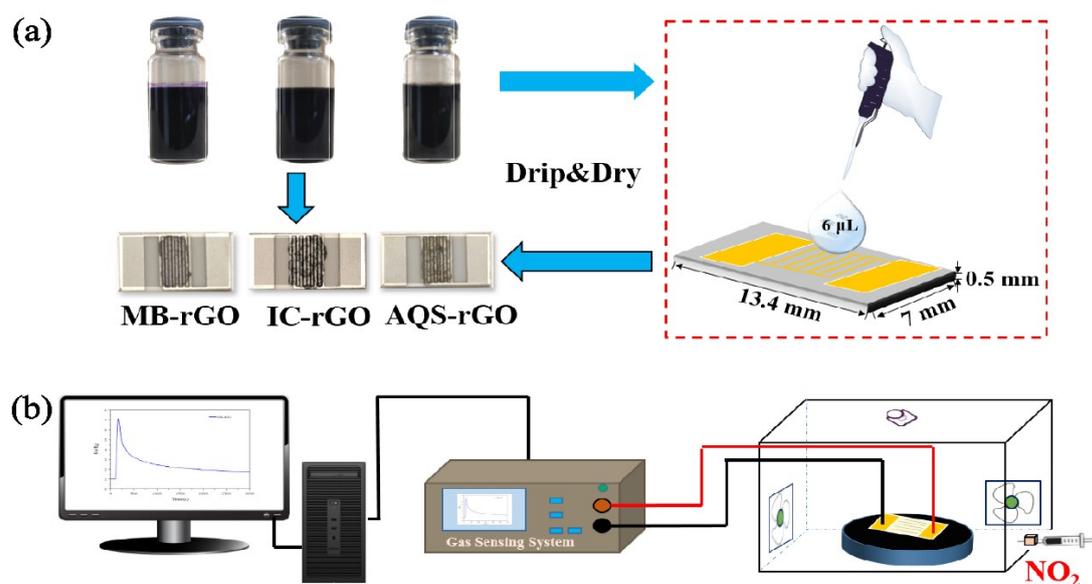


Figure S7. Schematic diagram of (a) the preparation of typical graphene-based gas sensors and (b) the home-built gas sensing test system.

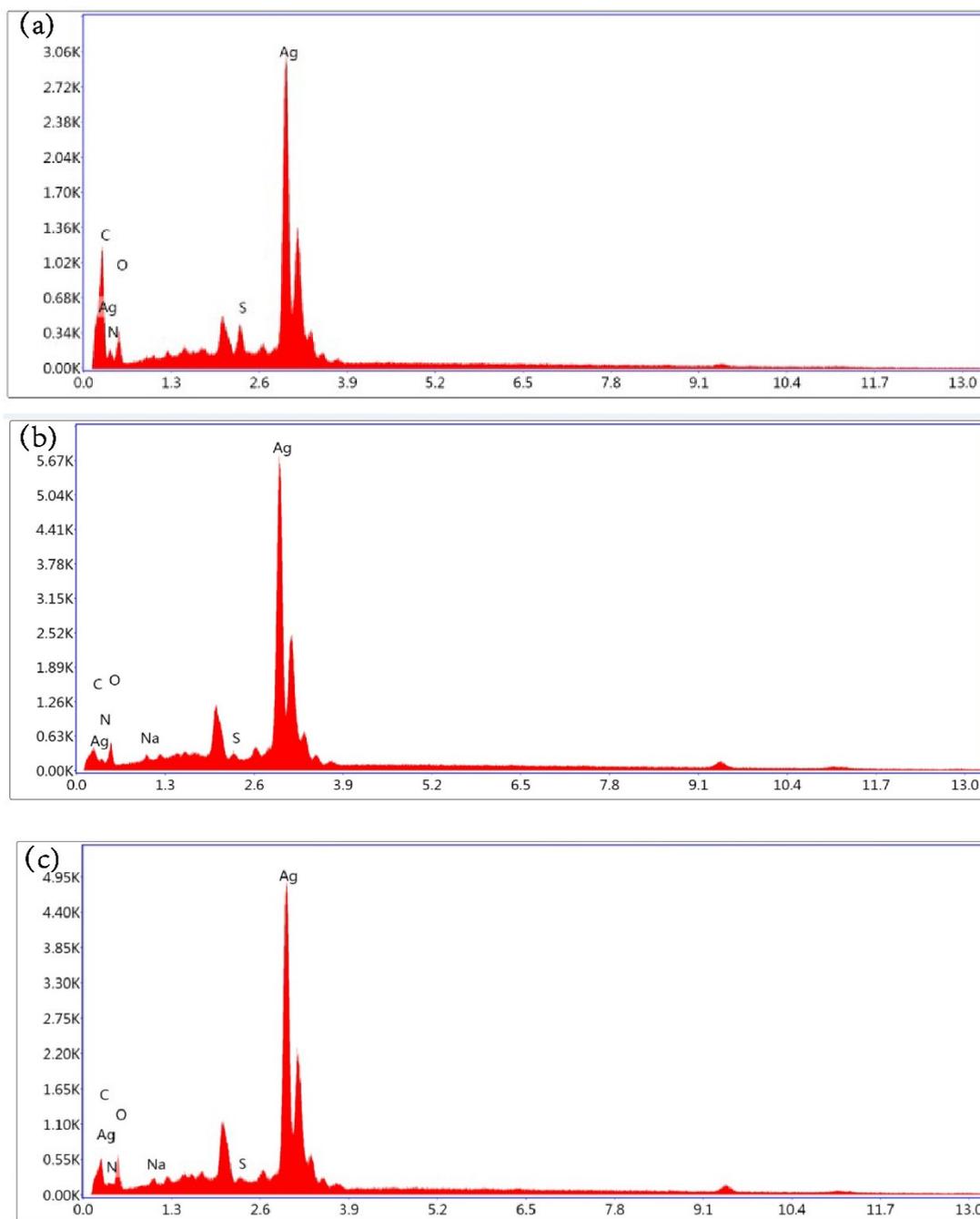


Figure S8. The EDS spectra of (a) MB- rGO, (b) IC- rGO and (c) AQS-rGO.

Table S1. The atomic content (%) of C, O, N, and S in GO, rGO, AQS-rGO, IC-rGO

and MB-rGO samples calculated by the XPS results.

Sample	C At%	O At%	N At%	S At%
GO	68.5	30.9	0.1	0.5
rGO	82.8	16.5	0.2	0.5
AQS-rGO	66.9	29.9	0.2	3.0
IC-rGO	59.7	32.6	4.5	3.2
MB-rGO	80.4	11.3	5.2	3.1

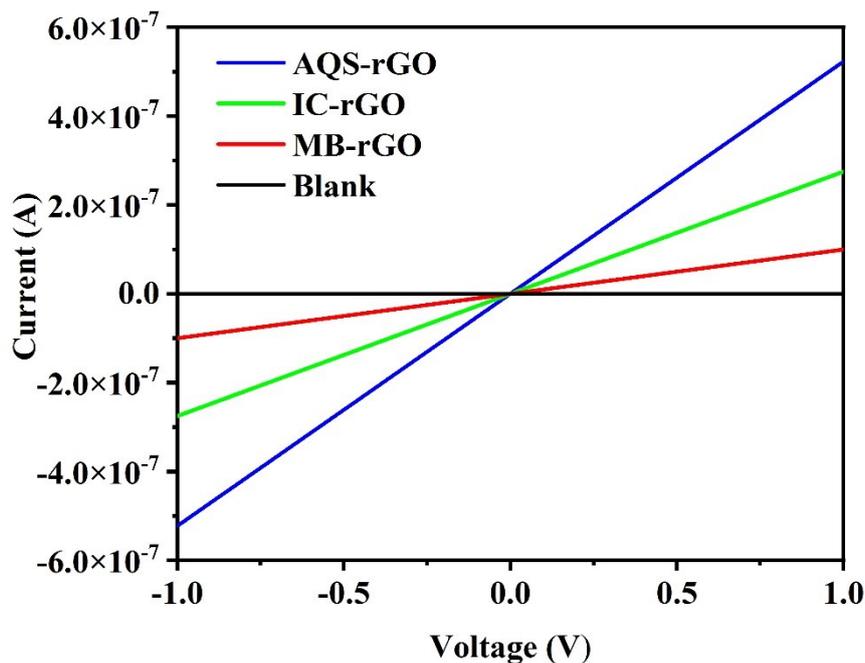


Figure S9. Current versus voltage curves of the blank, MB-rGO, IC-rGO and AQS-rGO sensor.

The relationship between current and voltage (I-V) was linear from -1 V to 1 V, showing good ohmic contacts between the gas sensing materials and IDEs (Figure S9). In other words, there is no Schottky barrier between these three graphene-based gas sensing materials and IDE.

Table S2. Electrochemical properties of the three molecules

Molecule	λ_{onset}	$E_{\text{g}}^{\text{opt[a]}}/\text{eV}$	E_{ox}/V	HOMO/eV	LUMO/eV
MB	745	1.66	0.21	-4.61	-2.95
IC	710	1.74	-0.01	-4.39	-2.65
AQS	380	3.26	-0.12	-4.28	-1.02

[a] $E_{\text{g}}^{\text{opt}}$ was calculated from $1240 \text{ nm}/\lambda_{\text{onset}}$, [b] $\text{HOMO} = - (E_{\text{ox}}^{\text{onset}} + 4.80 - E_{\text{Fc}/\text{Fc}^+}) \text{ eV}$; [c] $\text{LUMO} = \text{HOMO} + E_{\text{g}}^{\text{opt}}$.

1. Choi, C. H.; Park, S. H.; Woo, S. I., Heteroatom doped carbons prepared by the pyrolysis of bio-derived amino acids as highly active catalysts for oxygen electro-reduction reactions. *Green Chemistry*, 2011, 13, 406-412.